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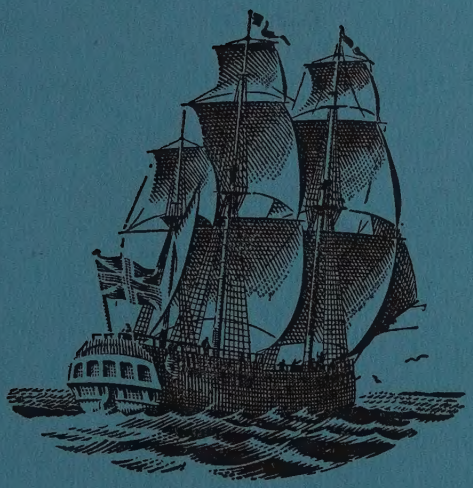

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The British quarterly scientific journal ENDEAVOUR was first published, by Imperial Chemical Industries Limited, in January 1942. Its purpose is to provide scientists, especially those overseas, with news of the progress of the sciences. While emphasis is laid upon British work, narrow insularity is avoided by publishing a number of articles from overseas contributors and by impartial reference to the world's scientific literature. To make the journal truly international in character it is published in five separate editions—English, French, German, Italian, and Spanish.

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The drawing on the cover is of the bark Endeavour, *which, commanded by Captain James Cook and carrying a number of scientific workers, was sent out by the British Admiralty in* 1768 *to chart the South Pacific Ocean and observe the transit of Venus*

ENDEAVOUR

A quarterly review designed to record the progress of the sciences in the service of mankind

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Joseph Black

Joseph Black had little interest in publication, preferring to communicate his knowledge by word of mouth, and it is perhaps not generally appreciated that his deservedly great reputation as an experimental chemist rests virtually on one published paper—'Experiments upon Magnesia Alba, Quicklime and some other Alcaline Substances'. But this one masterly paper, in which he quantitatively interprets the relationship between carbon dioxide (fixed air) and magnesia, lime, and other simple alkalis, has now stood the test of two centuries—for his final account of his experiments was read in June 1755. This paper may be justly said to mark the beginning of a new era of chemistry, in which the balance became the arbiter of experiment. The immensely fruitful results which stemmed from the introduction of quantitative methods need no emphasis now.

Black was born, one of a family of thirteen children, at Bordeaux in 1728. His father was an Irish merchant, and his mother, who undertook his early education, a Scotswoman. At the age of fourteen he was sent to Glasgow to study medicine at the university. There he came under the influence of Cullen, who was then professor of medicine and soon began, according to the custom of the time, to give lectures in chemistry, a subject not previously taught in Glasgow. Black was fortunate in attracting the favourable attention of Cullen, who not only made him assistant in his laboratory—a most valuable training in itself—but treated him as one of his own family.

Chemistry at that time was commonly looked upon as no more than a useful adjunct of pharmacy and had still not thrown off the mantle of medievalism. The teachings of Boyle had gone largely unheeded for two generations and the four Aristotelian elements were still generally accepted. Nevertheless, some unifying concepts had been developed. The phlogiston theory, which Black himself did not finally reject until 1791, deserves more respect than is sometimes accorded it; although it finally proved fundamentally wrong, it served for a long time as a stimulating working hypothesis and made possible some helpful classification of existing chemical knowledge. At the same time, the concepts of acids, bases, and salts were beginning to be formulated. The conversion of limestone into quicklime by strong heating was well known; in this process some innate virtue of the fire was supposed to be passed on to the lime. That mild alkalis could be made caustic by treatment with quicklime was ascribed to the passing on of this same fiery or caustic element. The true relationship between the various alkaline substances, which Black so brilliantly elucidated, was, however, still unknown.

Black's interest in this branch of chemistry was originally medical. Stone in the kidney was a well known but—in the then primitive state of surgery—very intractable condition, and attempts were made to dissolve and disperse the stones by treating them with caustic alkali. The dire effects likely to result from this kind of treatment need no description in the light of present knowledge, and it is not surprising that Cullen turned his thoughts to the possibility of using milder alkalis for the purpose; in consequence Black began a study of the properties of magnesia alba (magnesium carbonate). This substance had previously been studied by Friedrich Hoffmann, who distinguished between the oxides of calcium and magnesium. Black showed that the carbonates of the two metals differed in that on treatment with sulphuric acid one gave an insoluble product, whereas the other yielded a soluble substance which proved to be identical with Epsom salt.

Black's experiments on magnesia began in 1752, in which year he changed his place of study to Edinburgh. Two years later he presented for the degree of Doctor of Medicine a thesis entitled *De humore acido a cibis orto et magnesia alba* ('On the acid humour arising from food and on magnesia alba'). This thesis is a catalogue of thirty-two experiments, prefaced by some account of the nature of the digestive process. On his own admission, however, the latter was added principally to give the thesis the requisite medical flavour. When he read a revised version of the thesis in Edinburgh in the following year the study of digestion was omitted, but a second part, dealing at length with the constitution of chalk and lime, was added.

The differences between the versions of 1754 and 1755 are striking. Both reveal the essential facts, but a comparison reveals the man. It is a commonplace that study of the evolution of some work of art from sketch to sketch may lay bare the workings of the creative mind, but the history of science has hitherto been little studied from this point of view. Black's papers suggest that such study may be well rewarded. In 1754 Black expresses his

ideas about fixed air cautiously and with reservations. He makes tentative suggestions and describes isolated results: clearly he is reluctant to commit himself. But by 1755 all his doubts are resolved and, with brilliant logic, he expresses his ideas definitely and incontrovertibly. This paper is now acknowledged as a classic, and the discoveries described are today an essential part of chemistry.

The most important experiment was that in which a known weight of magnesia alba was calcined, the product was dissolved in acid, and the original weight of material was recovered by addition of mild alkali (sodium or potassium carbonate). Black concluded that the loss of weight on calcining was due to the loss of an 'air' normally 'fixed' in the magnesia alba and consequently 'fixed' also in the mild alkali. Similar experiments, described in the paper of 1755, he interpreted as showing that the formation of lime from chalk was due to the liberation of 'fixed air' and that the causticization of mild alkalis was due to the transference of 'fixed air', and not some imponderable caustic or fiery virtue, from substance to substance.

Black's work had many consequences, but the most important was that it showed the need to put the whole of chemistry on a quantitative basis —the balance became an indispensable laboratory tool. Moreover, by weighing gases in combination, Black brought these elusive substances, too, within the scope of quantitative work, and so made the first steps which prompted the giant strides of Priestley, Cavendish, and Lavoisier.

But this was not all, as Lavoisier saw clearly. The weights of reactants and products meant no more than any other physical properties unless there were some underlying unity. Only if matter is conserved can the balance make sense, and it is the cyclic quality of Black's experiments which is so significant. When Lavoisier came to define the role of oxygen in combustion his work with mercury had the same cyclic character as Black's.

As has been said, Black's reputation as a chemist rests largely, so far as published work is concerned, upon a single paper. He made, however, one other notable contribution to science, again while still a comparatively young man, but this was in the field of physics. The nature of heat was still not understood—and nearly a century was to elapse before the notion of heat as an intangible fluid, or caloric, was abandoned—but Black's ideas were in advance of his time. He was the first to understand the nature of latent heat, and in doing so he devised valuable new methods of calorimetry. He was aware of the practical implications of these investigations, and he lent James Watt fifteen hundred pounds to assist him in the development of the steam engine. Black's work on latent heat led Watt to realize that unnecessary condensation in the cylinders of his early steam engines caused a serious lowering of efficiency. Watt overcame the difficulty by effecting the condensation in a separate vessel and maintaining the temperature of the cylinder by means of a steam-jacket. Black then gave him further assistance by introducing him to John Roebuck, the ironmaster, who materially helped the further development of the 'condensing' engine, though it was some years before other technical difficulties, notably the accurate boring of cylinders, were overcome.

Black's experimental work marked the opening of a new era, and entitles him to be looked upon as one of the founders of modern chemistry, but neither chemistry nor science as a whole advances only through great discoveries. The teaching of science—the handing on of new knowledge, its understanding simplified by the formulation of unifying theories and the removal of errors and unnecessary detail—is no less important. In this sphere alone Black would have made a great name for himself, for he was certainly in the same class as great formative teachers like Lémery and Rouelle. His clear instruction and unhesitating leadership put a whole generation of chemists—among them Thomas Beddoes, 'discoverer' of Humphry Davy—in his debt. Never slow to acknowledge the work of others, he was among the first to teach the new doctrine of Lavoisier. It is quite untrue that Black clung unduly long to the phlogiston theory because of personal antipathy to Lavoisier.

In a dissertation of 1787 John Carmichael epitomizes the qualities of Black and Lavoisier, referring to the former as '*clarissimus*' and the latter as '*hodie primarius scientissimus*'; with this relative assessment most historians of chemistry will agree.

Alchemy in medieval Islam

E. J. HOLMYARD

For the period with which this article deals, any attempt to differentiate between alchemy and chemistry would be vain; chemistry as a science is not yet three centuries old, and the practical operations out of which it grew were the operations of alchemy. In the early Middle Ages the chief exponents of alchemy were Arabic-writing Muslims of various races, and our words alchemy and chemistry, whatever their ultimate origin, are derived immediately from the Arabic *al-kīmīa*. Western alchemy is almost entirely a direct legacy from Islam.

It is not yet possible to write a definitive account of alchemy in medieval Islam, for thousands of Arabic manuscripts on the subject lie unexamined in the great libraries of Europe, Egypt, Turkey, India, and Pakistan. Not until they have been studied can a final estimate be made, and there are far too few scholars engaged on this task to make its early accomplishment feasible. In spite of Berthelot's pioneer efforts towards the end of last century, little was known with certainty of Muslim contributions to alchemical theory and practice until the last few decades, though tradition, on the principle *omne ignotum pro magnifico*, ascribed to the Arabs a virtuosity in the occult arts that 'The Thousand Nights and a Night' subsequently did much to reinforce. At the present time, owing mainly to the researches of H. E. Stapleton, Julius Ruska, M. Plessner, and Paul Kraus, we can form a picture of Muslim alchemy that seems likely to be correct in outline, even though still seriously lacking in detail.

The Islamic era or Hegira dates from A.D. 622, the year of the Prophet's flight (*hijra*) from Mecca. One hundred and ten years later, a Muslim army was defeated at Poitiers by Charles the Hammer. In the interval, Islam had become a vast empire stretching from the Indus to the Pyrenees, and many and various races had become incorporated into its civilization. Some of these peoples continued to speak their own languages, but Arabic was the religious, official, and literary language throughout the empire. Thus it happened that numerous Muslim works on alchemy are Arabic only linguistically, their authors being of Persian or other nationality. The contribution of the Arabs to knowledge was no greater than that made by their Muslim, Christian, and Jewish subjects.

ORIGINS AND TRANSMISSION OF ALCHEMY

At the time of the rise of Islam, alchemy was already an art of venerable age. It was of a dual character. On the one hand it embraced specialized portions of metallurgy, dyeing, and similar crafts, and impinged at many points on medicine. On the other hand it was a semi-mystical cult of an eclectic kind, drawing its tenets whence it listed. The main aims of the alchemists were to prepare the philosophers' stone, which should convert base metals into gold, and further to prepare an elixir to prolong human life indefinitely.

The origin of alchemical theory is still far from clear, and possibly such speculation arose independently in more than one early civilization. In the west it certainly owed much to Aristotle's views on the nature of matter and to the Pythagorean theory of numbers [1], but the weight of evidence is that the fundamental ideas arose in the ancient Persian Empire, which included Mesopotamia, Asia Minor, Syria, and Egypt. Forbes [2] suggests that three streams of thought contributed to the rise of alchemy:

(*a*) The philosophy and technology of the ancient Near East.

(*b*) The philosophical tenets of the Persian and Indian civilizations.

(*c*) The philosophy and science of the Greeks.

The earliest alchemical treatises come from Hellenistic Egypt. Perhaps the most ancient of them is the *Physika kai Mystika*, 'Physical and Mystical Matters', of an author writing under the name of Demokritos; he probably lived in the second century A.D. and is not to be confused with the celebrated philosopher Demokritos of Abdera (d 376 B.C.). A large collection of alchemical writings was made by Zosimos of Panopolis (*c* A.D. 300), who also wrote original works on the subject, and many commentaries on earlier authors were compiled by succeeding Graeco-Egyptians such as Olympiodorus (*c* 500?). Further north, alchemy was practised by the Sabians of Harran (*Carrhae*) in Syria. Harran, long since

destroyed, was a cosmopolitan city at which, during the Achaemenid period (*c* 650–530 B.C.), there began to take shape a remarkable intermixture of Persian, Syrian, and Greek natural philosophy. This syncretic system remained a characteristic of Harran for several centuries, and the city's reputation for catholicity of learning was still high in early Islamic days. Harran is also known to have possessed many able metallurgists, and to have traded extensively in the precious metals, sulphur, sulphides of arsenic, borax, and other substances commonly used in alchemy.

The Muslim conquerors soon manifested an enthusiasm for knowledge, and having overrun not only Alexandria and Harran but all the other principal centres of Greek learning they were able to indulge it to the full. Under rulers such as Hārūn al-Rashīd (764?–809) and Al-Ma'mūn (786–833) large numbers of academies and observatories were set up and the chief Greek works on philosophy, astronomy, mathematics, medicine, and other sciences were translated into Arabic, principally by Syriac-speaking Nestorian Christians. From the eighth century onwards Islam was producing scholars of her own.

According to Al-Nadīm, a tenth-century biographer, the first Muslim to interest himself in alchemy was Khālid ibn Yazīd (d 704), a son of the royal house but never himself Commander of the Faithful. Stapleton [3] renders the relevant passage as follows:

> Being himself a scholar, he [Khālid] was greatly interested in the sciences. He was particularly attracted to the Art [of *Kīmīa*]: so he ordered some Greek philosophers who lived in the city of Misr [Cairo], and who had a good knowledge of Arabic, to be summoned, and instructed them to translate the books on the Art from the Greek and Coptic languages into Arabic. This was the first translation from one language to another in Islam.

Some Arabic alchemical works ascribed to Khālid are still in existence but have not yet been closely studied; most of them, like those of many later alchemists, are in verse.

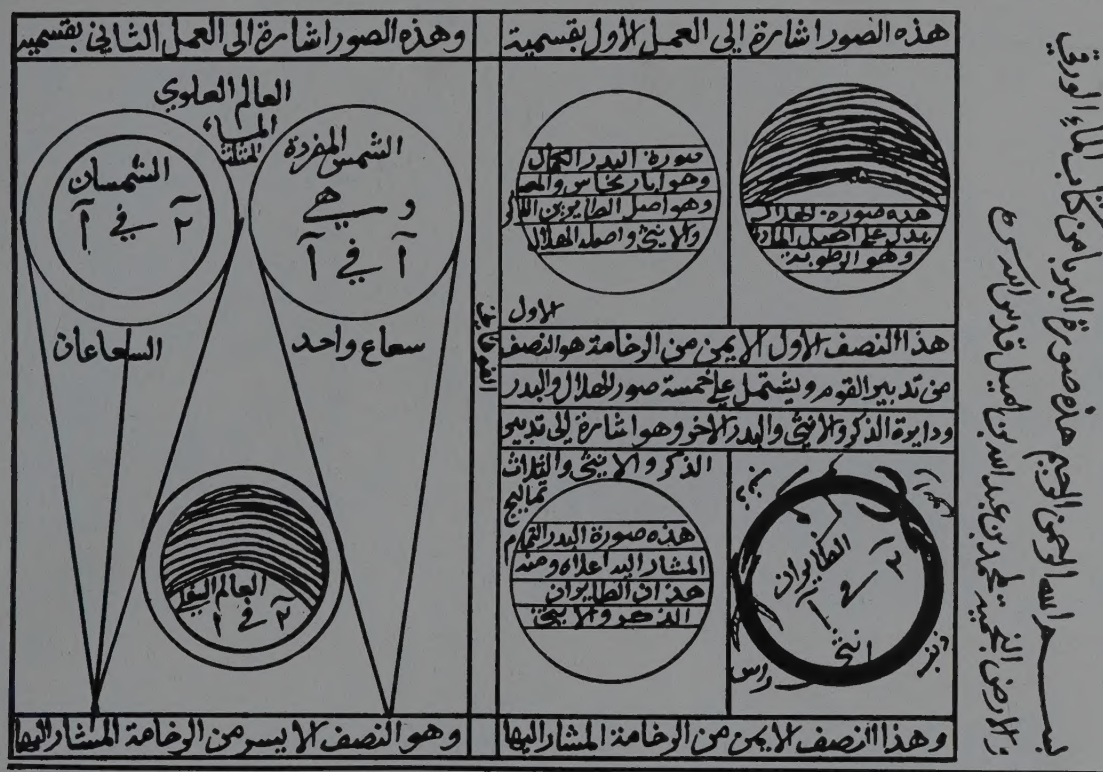
JĀBIR IBN ḤAYYĀN

The greatest name in alchemy, whether Muslim or Christian, is that of Jābir or Geber: alchemists themselves were described by one of their number, Thomas Norton of Bristol (fifteenth century), as 'Geber's cooks'. The life and works of this master of the art have presented a very difficult historical problem, which, however, research carried out during the last fifteen years has gone far to solve. There seems no reason to doubt that at the court of Hārūn al-Rashīd there was an alchemist named Jābir ibn Ḥayyān al-Ṣūfī, the son of a druggist of Kufa. As a young man he enjoyed the patronage of the sixth Shī'ite Imam, Ja'far al-Ṣādiq, and later that of the caliph's powerful ministers the Barmecides. He was probably born in 721 or 722, but the date of his death is unknown; he is said to have survived the banishment of the Barmecides in 803. Muslim chroniclers describe him as a man of culture and scholarship, and say that he was active in securing the importation of further Greek works on science and philosophy from Byzantium.

Difficulties begin to arise when consideration is given to the very numerous treatises on alchemy that pass under his name. There are so many of them that it seemed hardly possible for one man to have written them all, and the suspicion grew that some at least were fathered on him by later writers. The brilliant investigations of Kraus [4] showed that the suspicion was well founded. Critical examination of the Jābirian Arabic *corpus* proved beyond doubt that much of it must have been recensed or perhaps originally written by members of the Ismā'ilite sect early in the tenth century. How much of the alchemical operations and theory described in it is of the eighth century and how much of the tenth must remain a matter of conjecture: here, for economy of words, it will be referred to merely as 'Jābir'.

On the constitution of matter, Jābir held the Aristotelian conception of the four elements, fire, air, water, and earth, but developed it on different lines. He postulated first the existence of four elementary qualities or 'natures', namely hotness, coldness, dryness, and moistness. When these natures united with substance they formed compounds of the first degree, namely hot, cold, dry, moist. Union of two of these gave rise to fire (hot + dry + substance); air (hot + moist + substance); water (cold + moist + substance); and earth (cold + dry + substance). In metals, two of the 'natures' are external and two internal, a point to which further reference is made later (p. 122). Thus in his 'Book of Seventy' Jābir says that lead is cold and dry externally and hot and moist internally, as is also silver (but see p. 122); gold, on the other hand, is hot and moist externally and cold and dry internally.

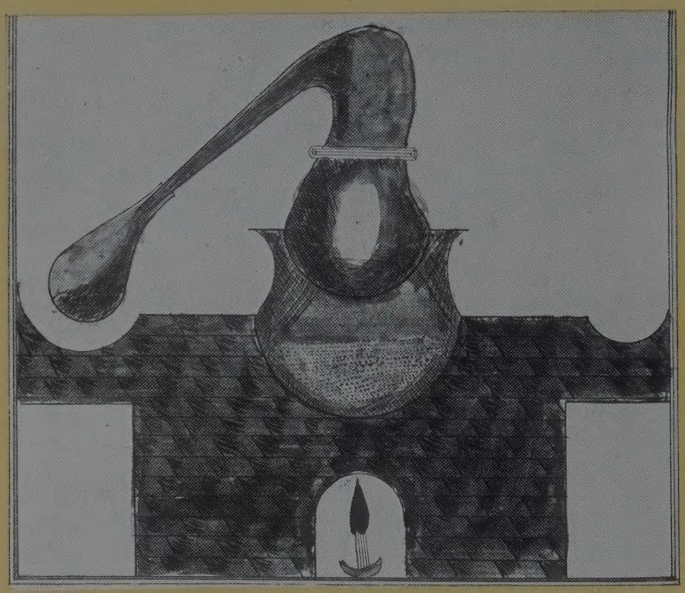
He believed that, under the influence of the planets, metals were formed in the earth by the

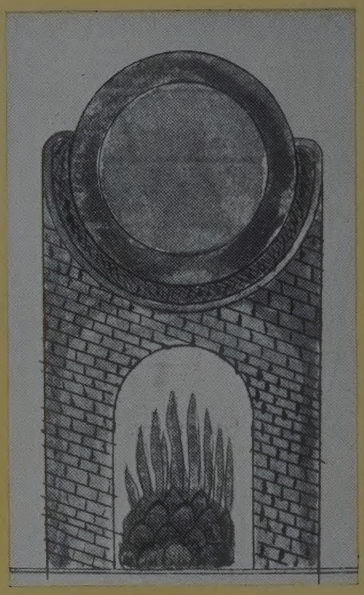


And these pictures indicate the 'Second Operation' in its two parts.	These pictures indicate the 'First Operation' in its two parts.
The Higher World The Triple Water (i) The Two Suns, Two in One The Two Rays The Single Sun, and it is One in One Single Ray The Lower World, Two in One	The picture of the Full Moon, and it is the *Abār-Nuḥās* and *Magh(nīsiyā)*, and it is the origin of the two birds, male and female, and its origin is the New Moon. This is the picture of the New Moon, indicating the origin of matter (*Aṣl al-Māddah*), which is moisture. This is the first (and the) right half of the tablet. It is the first half of the 'Operation of the People' and includes five pictures—the New Moon, the Full Moon at its zenith, the Circle of the Male, the Circle of the Female, and another Full Moon. And this indicates the 'Operation of the Male and the Female' and the 'Three Saltings.' This is the picture of the Perfect Full Moon, referred to above, and from this come these Two Birds, the male and the female. Tail Male Head The Two Birds Two in One Head Female Tail
And this is the left half of the marble tablet referred to above.	And this is the right half of the marble tablet referred to above.

Bismillāh. This the Pyramid (*Birbā*) Picture from the book called *Al-Mā' al-Waraqī wa'l Ard an-Najmiyah* ('Silvery Water and Starry Earth') by Muhammad bin 'Abdullāh bin Umail. May Allāh sanctify his heart!

FIGURE 1 – *Facsimile of a page of an Indian MS of the Mā' al-Waraqī containing symbolical pictures (with translation of texts given below).* (*From* Mem. Asiat. Soc. Beng., **12**, No. 1, *Plate* 1, 1933.) *(By courtesy of Dr H. E. Stapleton.)*





FIGURES 2 and 3 – *Stills, from an Arabic alchemical manuscript* (British Museum Add. 25724).

FIGURE 4 – *From an Arabic alchemical manuscript. The inscription reads: Know that the compound referred to has specific weights that should be in equilibrium, so that the hotness does not overcome the coldness or the dryness overcome the moistness. For what is in equilibrium will possess permanent natures and will never undergo change, while natures not in equilibrium will suffer alteration.* [*Sun to the left, Moon to the right. Figures between possibly represent iron, copper, lead, tin, mercury, and Khār ṣīnī.*] (British Museum Add. 25724.)

union of sulphur (which would provide the hot and dry 'natures') and mercury (providing the cold and moist). This theory, which appears to have been unknown to the ancients, may have been original, though it is possible that Jābir appropriated it from Balāniūs (Apollonius of Tyana). The reason for the existence of different kinds of metal is that the sulphur and mercury are not always pure, and that they do not always unite in the same proportion. If they are perfectly pure, and if also they combine in the most complete natural equilibrium, then the product is the most perfect of metals, namely gold. Defects in purity, and particularly in proportion, result in the formation of silver, lead, tin, iron, or copper, but since these metals are essentially composed of the same constituents as gold, the accidents of combination may be rectified by suitable treatment. Such treatment is the object of alchemy, and is to be carried out by means of elixirs.

Jābir was convinced that to try to effect transmutations empirically was a waste of time; he believed that order reigned in the material world, and that qualitative changes in substances could be explained on a quantitative basis. He was thus led to his characteristic conception of 'balance' (*mīzān*), which he elaborated particularly in his 'Books of the Balance' (*Kutub al-Mawāzīn*). By balance he did not refer to equality of weight; in fact, although he does describe a hydrostatic balance, the importance of comparing masses was as little perceived by him as by all other alchemists. His balance was an equilibrium of elementary qualities or 'natures', and a great deal of his work was devoted to attempts to establish the equilibrium figures in gold—the perfect metal—so that the same balance could be effected in base metals, thus bringing about their transmutation.

Except for the history of ideas, it would be unprofitable to follow the development of such a scheme in detail, but it is of interest to examine one of the lines of attack. This has been done by Kraus [5] and Stapleton [6], on whose work the following account is principally based. We find throughout Jābir's works a series of numbers to which he attaches great importance: it is 1, 3, 5, 8 (totalling 17), and 28. He says that everything in the world is governed by the number 17—metals, for instance, have 17 'powers'. Now the numbers composing the total of 17, namely 1, 3, 5, 8, recall the magic square of the first nine digits:

4	9	2
3	5	7
8	1	6

Here the total is 45, but analysing the square gnomonically, as indicated by the heavy lines in the figure, gives 1, 3, 5, 8 in the remaining square, while the total in the gnomon is 28. This magic square, which was known to the Neoplatonists of the third century and is doubtless much older, is thus clearly the source of Jābir's significant numbers. He does not appear to have specifically mentioned it, but it must have been familiar to him not only from his acquaintance with Neoplatonist writings but because it had significance in the Ṣūfī mystical society of which he was a member.

One way in which Jābir used these numbers was in the application of alphabetical numerology to elucidate the constitution of the metals. Each of the four elementary qualities or 'natures' was supposed to have four degrees and seven subdivisions, giving a total of 28×4 'positions'. The letters of the Arabic alphabet, 28 in all, were assigned to the subdivisions of heat, coldness, dryness, and humidity, and the scheme was extended to the values of the four degrees according to the series 1, 3, 5, 8. The degrees and subdivisions were equated to weights on the Arabic system of 2 *qīrāṭs* = 1 *dānaq*, 6 *dānaqs* = 1 *dirham*, and a table was constructed in which, for example, the letter *b* denoted, in the second degree of coldness, a weight of $3\frac{1}{2}$ *dirhams*; in the fourth degree *b* corresponded to a weight of $9\frac{1}{3}$ *dirhams*. The remaining letters were similarly 'calibrated'.

To determine the 'balance' of lead on this scheme, its name was analysed arithmo-alphabetically, only the consonants being used. Lead in Arabic is *usrub*, but the first letter, not indicated in the transliteration, is the consonant *alif*. *U* being a vowel, the operative letters are therefore *alif*, *s* (*sīn*), *r* (*rā*), and *b* (*bā*). *Alif*, being the first letter of the name, represents heat of the first degree, and is equivalent to a weight of 7 *dānaqs*; *sīn*, the second letter, represents dryness of the second degree, and is equivalent to 1 *dirham*; *rā* represents humidity of the third degree, $1\frac{1}{2}$ *dirhams*; and *bā* represents coldness of the fourth degree, that is $9\frac{1}{3}$ *dirhams*. Hence a lump of lead weighing $12\frac{3}{4}$ *dirhams* would contain the above weights of heat, dryness, humidity, and coldness, and this

composition would hold for any specimen of lead. The fact that another name for lead, namely *raṣāṣ*, gives an entirely different result may cause misgivings as to the reliability of this method of analysis.

It was mentioned earlier that Jābir distinguished between the external and the internal composition of a metal. One reason for this distinction can be found in the figures just elicited. Metals are composed of heat, cold, dryness, and humidity, but there is a limiting condition: opposing 'natures' are in the ratio of either 1 : 3 or 5 : 8 or *vice versa*. The figures for lead, however, do not agree with these ratios, and the difficulty is even greater when *fiḍḍa* (silver) is analysed in the same way—it proves to consist merely of heat and coldness in equal proportions. Jābir was therefore forced to use a further hypothesis, namely that the analysis reveals only the peripheral constitution; the balance must be restored by the constitution of the interior. Hence for silver the total composition, external and internal together, must be arrived at by calculation:

Heat:
$1\frac{1}{8}$ *dānaqs* + a complement of $5\frac{1}{8}$ *dānaqs*
$= 1\frac{1}{8}$ *dirhams*

Coldness:
$1\frac{1}{8}$ *dānaqs* + a complement of $3\frac{1}{8}$ *dirhams*
$= 3\frac{1}{8}$ *dirhams* $(= 3 \times 1\frac{1}{8})$

Humidity:
0 *dānaqs* + a complement of $5\frac{5}{8}$ *dirhams*
$= 5\frac{5}{8}$ *dirhams* $(= 5 \times 1\frac{1}{8})$

Dryness:
0 *dānaqs* + a complement of $9\frac{1}{8}$ *dirhams*
$= 9\frac{1}{8}$ *dirhams* $(= 8 \times 1\frac{1}{8})$

$19\frac{1}{8}$ *dirhams* $(= 17 \times 1\frac{1}{8})$

The unit of $1\frac{1}{8}$ seems to be chosen here so that fractions of a *dirham* not represented by either a *dānaq* or a *qīrāṭ* are avoided.

The transmutation of one metal into another is thus an adjustment of the ratio of the manifest and latent constitutions of the first to those of the second, an adjustment to be brought about by an elixir. According to Jābir there are various elixirs suitable for specific transmutations, but transmutations of every kind can be brought about by a grand or master elixir (*al-iksīr al-aʿẓam*). The Alexandrian and Harranian alchemists appear to have used only mineral substances in their attempts to prepare elixirs for transmutation, but Jābir was an innovator and introduced both animal and vegetable products to the alchemical armoury. Among the former he mentions the marrow, blood, hair, bones, and urine of lions, vipers, foxes, oxen, gazelles, and donkeys both domesticated and wild. Suitable plants included aconite, olive, jasmine, love-in-a-mist, onion, ginger, pepper, mustard, pear, and anemone.

Such lists provide an indication that Jābir was more than a theorizer, and there is indeed much in his books to show that he was well versed in chemical operations. Though his theory is complex, obscure, and often to our modern minds ridiculous, he can be perfectly clear when giving instructions for a preparation. A single example must suffice:

> To convert mercury into a red solid. Take a round glass vessel and pour a convenient quantity of mercury into it. Then take a Syrian earthenware vessel [Syrian pottery enjoyed a high reputation in those days] and in it put a little powdered sulphur. Place the glass vessel on the sulphur and pack it round with more sulphur up to the brim. Place the apparatus in the furnace for a night, over a gentle fire . . . after having closed the mouth of the earthenware pot. Now take it out and you will find that the mercury has been converted into a hard red stone the colour of blood . . . it is the substance that the learned call cinnabar.

Of alchemical equipment, both substances and apparatus, Jābir gives a less systematic description than Rāzī (p. 123), but he does classify minerals into three groups. These are (*a*) spirits, or substances that volatilize completely on heating; (*b*) metals, or fusible substances that are malleable, sonorous, and possess a lustre; and (*c*) substances that, whether fusible or not, are not malleable and may be powdered. The spirits numbered five: sulphur, arsenic [sulphides], mercury, camphor, and sal ammoniac. This is one of the earliest Arabic mentions of sal ammoniac (*nūsẖādur*), which for a time was imported from inner Asia; it was probably obtained there as a sublimate from burning coal-seams [7]. Jābir, however, knew how to prepare it from organic matter, and distinguishes between the mineral form and the sal ammoniac 'from hair'.

There were seven metals, namely gold, silver, lead, tin, copper, iron, and *kẖār ṣīnī*. The last of these has not been identified with certainty; the name signifies 'Chinese iron', and Muslim writers say that it was used in China to make mirrors with the power of curing ophthalmic maladies of sufferers looking into them. It was also cast into bells of a particularly melodious tone. According to Laufer [8], it was an alloy chiefly composed of

copper, zinc, and nickel; it gave a silvery surface when polished and was known as *pai-t'ung* or white copper. There could have been very little of it available in Islam, and for the most part it was included among the metals only to bring their number up to seven when mercury was classed as a spirit.

Jābir's classification of minerals other than spirits and metals was not consistent, but in one book he sorts them into eight groups, according to whether they are (*a*) stony or not stony, (*b*) pulverizable or not pulverizable, and (*c*) fusible or non-fusible. Though such a system cannot be pressed very far, it is at least on sensible lines.

It is a matter of considerable interest that the Arabic text of the celebrated *Tabula Smaragdina*, or Emerald Table, of 'Hermes' was first discovered in Jābir's 'Second Book of the Element [στοιχεῖον] of the Foundation' [9]; before this discovery (1923) it had been known only in medieval Latin. Jābir says that he is quoting Apollonius of Tyana, and another version of the *Tabula* has been found in a book called 'The Secret of Creation' (*Sirr al-Ḫalīqa*) probably dating from the early ninth century and ascribed to Apollonius. The original language of the *Tabula* is unknown, but there are indications that it may have been Syriac. In its Latin form it is perhaps the best known piece of alchemical literature.

Some final remarks on Jābir must concern the problem of the authorship of the Latin works, such as the *Summa perfectionis*, ascribed to him. These appear first about the end of the thirteenth century, and of only one of them (the 'Book of Seventy'—not included in any of the collected Latin editions) is an Arabic original known. The others admittedly show signs of Muslim influence, but that is true of most early western alchemical works and is only to be expected in view of the fact that alchemy in Latin Europe was almost entirely an importation from Islam. The weight of evidence is in favour of the judgment that, because of the authority of his name, works written by an anonymous European alchemist were circulated as Jābir's.

RĀZĪ

Later than Jābir ibn Ḥayyān, though possibly contemporaneous with the writing of some of the Jābirian *corpus*, was Abū Bakr Muḥammad ibn Zakariyya, known as Al-Rāzī, 'the man of Ray' (*Rhagae*), from his birthplace near Teheran. He lived from 866 to 925. Rāzī was the greatest physician of the Middle Ages, and his works on medicine enjoyed an unquestioned authority extending over some six hundred years. He made many contributions to gynaecology and ophthalmology, and the monograph in which for the first time he distinguished between smallpox and measles is one of the classics of medicine. He practised at his native town and also at the great hospital in Baghdad, and his success as a clinician was matched by his outstanding ability as a teacher of medicine.

Like many physicians of medieval times, he was attracted to the study of alchemy and is said to have written a score of books on the subject; they have not all survived, but one of them, 'The Book of the Secret of Secrets' (*Kitāb Sirr al-Asrār*), has been translated into German and furnished with a commentary by Ruska [10]. Much of his alchemical work has also been studied by Stapleton, who places him on an intellectual level with Galileo and Boyle.

The metaphysical background of Rāzī's alchemy resembles that of the Jābirian writings: it is Ismā'ilite and therefore tends towards the spirit of free inquiry. The object of alchemy is still the transmutation of base metals into gold by means of elixirs, but a study of the *Kitāb Sirr al-Asrār* and other books of Rāzī conveys the impression that he was much more interested in practical chemistry than in theoretical alchemy. The *Kitāb Sirr al-Asrār* foreshadows a laboratory manual, and though the procedures described are often difficult to interpret they are probably representative of experiments that Rāzī himself had carried out. From the list he gives of materials and apparatus it is evident that his laboratory was very well equipped. It had beakers, flasks, phials, basins, glass crystallizing-dishes, jugs, casseroles, candle-lamps, naphtha-lamps, braziers, athanors, smelting-furnaces, files, spatulas, hammers, ladles, shears, tongs, sand- and water-baths, filters of haircloth and linen, alembics, aludels, funnels, and mortars. In addition, Rāzī gives details of the construction of more complicated pieces of apparatus from these and other units.

His store-cupboard contained not only specimens of all metals then known, but pyrites, malachite, lapis lazuli, gypsum, haematite, turquoise, galena, stibnite, alum, green vitriol, natron, borax, common salt, lime, potash, cinnabar, white lead, red lead, litharge, ferric oxide, cupric oxide, verdigris, and vinegar. Stapleton has adduced reasons for believing that Rāzī was also familiar with caustic soda and glycerol. It is uncertain whether he was acquainted with sulphuric and nitric acids,

but nitric acid was known to Jābir, who describes its preparation, and it was early used for the parting of gold and silver.

AVICENNA

Abū 'Alī ibn Sīnā, known to the west as Avicenna, was born in 980 and died in 1036–7. Like Rāzī, he was a Persian of extraordinary intellectual capacity and a prolific writer, and his books had wide popularity both in Islam and in medieval Europe. It has been well said of him that his thought represents the climax of medieval philosophy. His most important works were the 'Canon of Medicine' and the philosophical encyclopaedia *Kitāb al-Shifā'*, or *Sanatio*.

In the mineralogical section of the *Shifā'*, which in medieval times and in Latin translation circulated under the title *Liber de mineralibus* and was ascribed to Aristotle [11], Avicenna accepts the sulphur-mercury theory of metals but expresses emphatic disagreement with the claims of the alchemists. There is little doubt, he says, that alchemists can contrive to make solids closely resembling gold and silver, but these products are mere imitations. 'I do not deny', he proceeds, 'that such a degree of accuracy in imitation may be reached as to deceive even the shrewdest, but the possibility of transmutation has never been clear to me. On the contrary, I regard it as impossible, since there is no way of splitting up one metallic combination into another. Those properties that are perceived by the senses are probably not the differences that distinguish one metallic species from another, but rather accidents or consequences, the essential specific differences being unknown.' In other words, while accepting that metals were of a compound nature, Avicenna thought their structure too stable to be affected by fusion and other alchemical methods. 'It is likely', he writes, 'that the proportion of the elements that enter into the composition of each of the metals is different from that of any other. If this is so, one metal cannot be converted into another unless its structure is broken up and converted into the composition of that into which its transformation is desired. This, however, cannot be effected by fusion, which maintains the union and merely causes the introduction of some foreign substance or virtue.'

Avicenna's views received little support; they were in advance of their time, and the alchemists pursued their search for elixirs undisturbed by doubts and with undiminished enthusiasm.

LATER WRITERS

A younger contemporary of Avicenna was Ibn Umail, one of whose alchemical books, 'The Silvery Water' (*Al-Mā' al-Waraqī*), was translated into Latin in the Middle Ages and is printed in Zetzner's *Theatrum Chemicum* (1622). This work, the Arabic text of which was edited by Turab Ali [12] in 1933, is important as containing numerous quotations from earlier alchemical treatises otherwise unknown. Ibn Umail was of the opinion that the pyramids of Egypt were designed for the practice of alchemy, thus agreeing with the historian Al-Nadīm (*c* 988), who says: 'A pyramid contains rooms of different shapes in which are places for admixture, pounding, solution, coagulation, and distillation, which indicates that each of them was built for the art of *Kīmīa*.' Symbolic 'pyramid' figures from a manuscript of the *Mā' al-Waraqī* are reproduced in figure 1.

An early tenth-century work, ascribed—though with doubtful justification—to the Spanish Arab Maslama of Madrid, is called 'The Sage's Step' [13]. It contains the usual preponderance of theory, but there is also much straightforward information on various practical operations, particularly the refinement of gold and silver by cupellation and other methods. The author appears to have had more regard to quantitative observation than was usual, and in one experiment took a quarter of a pound of mercury, heated it moderately for forty days, and then weighed the resulting red powder; he could find no change in weight, but the fact that he made the determination is indicative of a careful and inquiring mind.

During the twelfth and thirteenth centuries the transmission of alchemical theory and practice from Islam to the west was accompanied by a decline in the number of Arabic writers on the subject: though perhaps future research will show that the decline was not as great as our present knowledge leads us to assume. Such books as were written were generally rearrangements of earlier ones or commentaries on them. Thus Abu'l-Qāsim al-'Irāqī, who lived in the thirteenth century, appropriated passages from the *Mā' al-Waraqī* and other books, with or without acknowledgment, and in doing so was merely following a common practice. There is, however, something to be said in favour of these purveyors of second-hand wares, for they often enable us to fill gaps in our knowledge of the earlier literature. One of them in particular, Aidamur al-Jildakī [14], is a most valuable source of information; with

him, this sketch of Muslim alchemy may fittingly end.

Little is known of Jildakī's life. He flourished in the first half of the fourteenth century, lived for a time in Cairo, and spent seventeen years in extensive travel to meet the principal alchemists of the time and to collect alchemical writings. Most of the remainder of his life he must have devoted to authorship, for he wrote at least twenty-five books, some of them very lengthy. Their value lies in the very numerous quotations that Jildakī fortunately saw fit to include in them, a value enhanced by the general accuracy with which the quotations are made. In many cases the original works are still in existence, and examination of them shows that Jildakī was a careful copyist; we may therefore with fair confidence accept as genuine other passages of which no earlier provenance is known.

The great bulk of Jildakī's work still awaits detailed study, but a full examination has recently been made by Taslimi [15] of the *Nihāyat al-Ṭalab* or 'End of the Search', which is a commentary by Jildakī on Abu'l-Qāsim al-'Irāqī's book known as the *Muktasab* [16]. The richness of the material offered by Jildakī may be judged from the fact that in the *Nihāyat* alone he quotes from, or mentions, no fewer than 42 works of Jābir and a large number of works by other authors, including Ibn Umail, Avicenna (whom he accuses of plagiarizing Al-Fārābī), Maslama, Khālid ibn Yazīd, and Rāzī. Another of Jildakī's books that would well repay study is the *Kitab al-Burhān* or 'Book of Proof', which contains a commentary on 'The Book of the Seven Idols' ascribed to Apollonius of Tyana. Taslimi says that there is a great deal of similarity between the ideas contained in the quotations from Jābir given in the *Nihāyat* and those found in the Latin works of Geber; he does not, however, think the correspondence sufficient to establish a close connection.

The author wishes to express his thanks to Dr Manuchehr Taslimi for the gift of a copy of the thesis mentioned in reference 15.

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Porphyrins

C. RIMINGTON

The porphyrins are among the most important of the natural pigments, but although they have been known for almost a century it is only within the last decade that the principal steps in their biosynthesis have been identified. This development is due in large measure to the introduction of isotopic labelling techniques. This research brings us towards an understanding of the biochemical lesions responsible for the various porphyria diseases.

Porphyrins are nitrogen-containing pigments which are very widely distributed in the animal and vegetable kingdoms. They occur either in the free state or in the form of complexes with metals, the iron complexes or haems being particularly important in animals, with which this article is primarily concerned. Much of our knowledge of the chemistry of the porphyrins we owe to Hans Fischer and his school at Munich, who from 1915 to 1945 carried out monumental researches upon their structure and chemistry. The first preparation of a porphyrin in a reasonable state of purity, however, was made much earlier; J. L. W. Thudichum, in a report to the British Privy Council in 1867 [1], described the action of strong acid upon blood and the separation from the mixture of a pigment which he called 'cruentine.' Some years later F. Hoppe-Seyler [2] renamed this substance 'haematoporphyrin', to indicate its red colour in solution and its origin from blood. This porphyrin is still known as haematoporphyrin and its chemical structure and relationship to haemoglobin are now clearly understood. Another most important pigment, still containing combined iron, was found to be readily prepared in crystalline form from haemoglobin by treating blood with hot acetic acid. Investigation of this substance, 'haemin', chiefly by Nencki, Küster, Willstätter, and finally by Fischer, revealed the true structure of the porphyrins.

The porphyrin molecule consists essentially of four pyrrole rings united by means of carbon bridges into a macrocyclic structure (figure 1). The simplest member would be that in which all the positions numbered 1 to 8 on the pyrrole nuclei were occupied by hydrogen atoms. This substance does not occur in nature, but can be prepared synthetically; to it Fischer gave the name 'porphin,' and on it he based a rational system of nomenclature for all the porphyrins. Thus, haematoporphyrin is known to have the structure shown in figure 2, and is accordingly described as 1, 3, 5, 8-tetramethyl-2, 4-di(α-oxyethyl)-porphin-6,

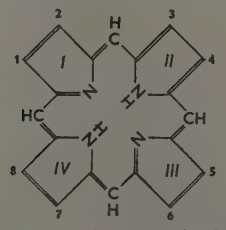


FIGURE 1 – *The porphyrin skeleton.*

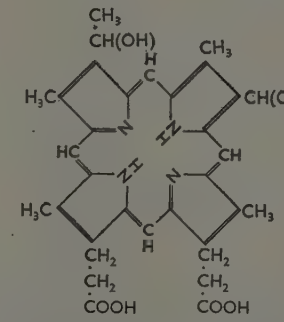


FIGURE 2 – *Haematoporphyrin.*

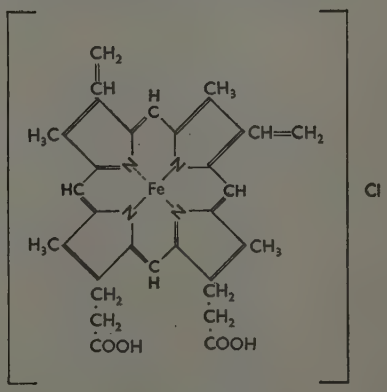


FIGURE 3 – *Haemin.*

7-dipropionic acid. Haemin (figure 3) is the chloride of an iron complex; the only difference from haematoporphyrin in the porphyrin structure is that positions 2 and 4 carry vinyl groups in place of hydroxyethyl groups. Removal of the co-ordinately bound iron from haemin leads to protoporphyrin (figure 4).

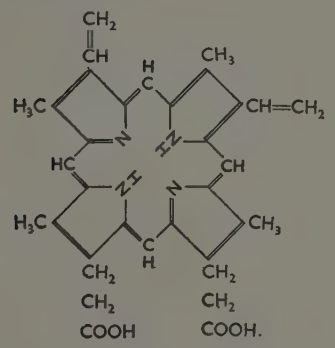


FIGURE 4 – *Protoporphyrin IX.*

It is clear that in a case such as that of protoporphyrin the formula depicted represents only one of the fifteen possible ways of arranging four methyl, two vinyl, and two propionic acid groups as substituents of the eight hydrogen atoms of porphin. The correctness of the structure reproduced, the ninth possibility, rests upon Fischer's synthetic work. Natural protoporphyrin is thus generally termed 'protoporphyrin IX.'

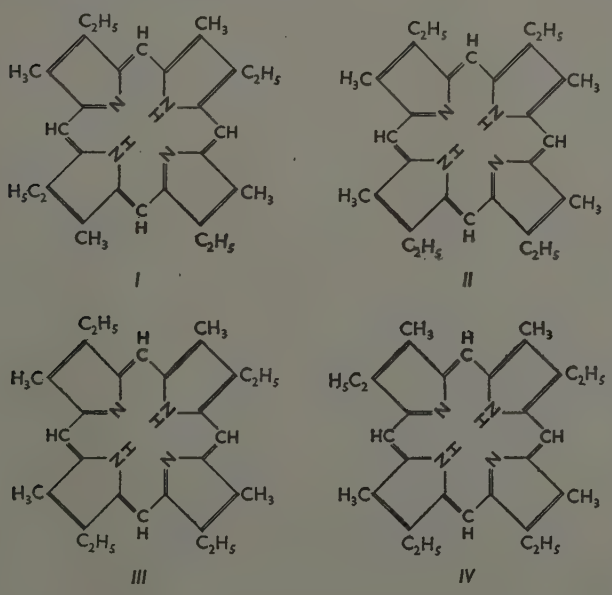


FIGURE 5 – *The four isomeric aetioporphyrins.*

Fischer also prepared synthetically all four possible isomers of aetioporphyrin, in which four methyl groups and four ethyl groups are present as substituents on the pyrrole rings (figure 5). Each aetioporphyrin can be regarded as the theoretical starting point of a series of porphyrins derived by inserting vinyl, acetic acid, propionic acid groups, etc., in place of the ethyl groups. Figure 6 illustrates the four coproporphyrins, the roman numeral indicating from which of the four aetioporphyrin isomers the pigment is theoretically derived. Protoporphyrin IX is seen to be a series III porphyrin (figure 4). Were derivatives of all four isomeric types produced by living organisms, the variety of natural porphyrins would be very great, but pigments of series II and IV do not appear to be formed by living cells; this limits the natural coproporphyrins (figure 6) and uroporphyrins (figure 7) to two in each case. Protoporphyrin IX seems to be the sole protoporphyrin to occur naturally.

It has already been stated that the porphyrins are widely distributed in nature. Their detection is rendered easy by the fact that in addition to colour, the highly resonating porphyrin ring structure confers upon them two other properties, namely the exhibition of well-defined and characteristic absorption bands in the visual and near ultra-violet regions of the spectrum, and of an intense red fluorescence in ultra-violet light which permits the detection of as little as one part of porphyrin in a thousand million parts of a suitable solvent. Normal urine contains some 10–200 micrograms per litre of a mixture of coproporphyrins I and III and a smaller quantity of uroporphyrin. About a quarter of a milligram of coproporphyrins and some protoporphyrin is eliminated daily in the faeces. Both coproporphyrin and protoporphyrin are present in the red blood cells, where the main pigment is of course haemoglobin, composed of the protein globin united with 'haem,' the iron complex of protoporphyrin IX. Iron porphyrin complexes combined with other specialized proteins constitute the cytochromes, the respiratory catalysts, which are present in every living cell which utilizes oxygen; catalase, an enzyme which rapidly destroys hydrogen peroxide; and similar catalysts vital to the life of the cell. Chlorophylls *a* and *b* are magnesium complexes of modified porphyrin structures.

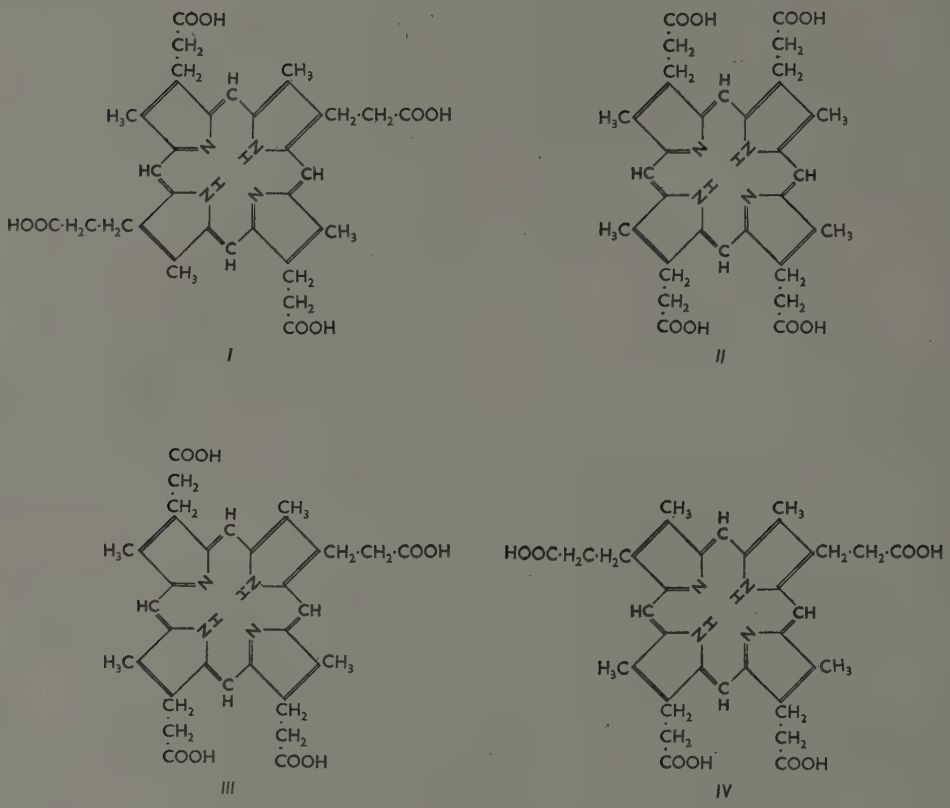


FIGURE 6 – *The four isomeric coproporphyrins.*

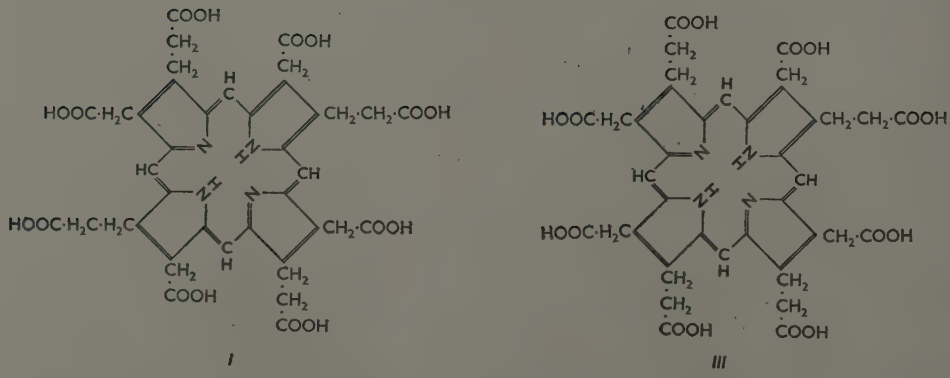


FIGURE 7 – *The two naturally occurring isomeric uroporphyrins.*

Free porphyrins, that is to say porphyrins not bound to metals, can be detected in small quantities in most animal tissues and organs, including parts of the nervous system, but in some species peculiar localizations occur. Thus in rodents, such as the rat, the gland of Harder, which is associated with the lachrymatory apparatus, contains large quantities of protoporphyrin; its appearance when viewed by filtered ultra-violet light is very striking (figure 9). Whereas the bony skeleton of most animals contains too little porphyrin to be directly visible, that of the ground squirrel (*Sciurus niger*) and of some other rodents has a distinctly brownish tint and under ultra-violet light exhibits a brilliant red fluorescence (figure 14), that of the teeth being particularly striking. Such an association of porphyrin with calcified tissue is met with frequently; other examples are the deposition of uroporphyrins in certain marine shells and in the zones of calcification of foetal bones. A unique occurrence of a porphyrin derivative in nature is that of the copper complex of uroporphyrin III in the flight feathers of the turacos (*Musophagidae*), African birds allied to the cuckoo, where it imparts a brilliant and very beautiful red colour to the wings.

We owe much of our knowledge of porphyrin biochemistry to the fact that certain rather rare disturbances of porphyrin metabolism occur both in man and in animals, and in these conditions excessive quantities of pigments are produced by the body. They are familial diseases. Thus in congenital porphyria, inherited as a Mendelian recessive character, there is from birth a large excretion in the urine and faeces of uroporphyrins and coproporphyrins, belonging mainly to series I, and the tissues of the body also become saturated with porphyrins. Since the porphyrins are strongly photoactive, this condition causes intense photosensitivity in the sufferers. On examination, the bones and teeth are seen to be reddish brown in colour, and under ultra-violet light they fluoresce brilliantly (figures 10, 15, 16, 17). In this disease freshly passed urine also is highly fluorescent because of the uroporphyrins and coproporphyrins it contains (figure 12). Another disturbance of porphyrin metabolism, termed porphyria cutanea tarda, shows many points of resemblance to congenital porphyria. Photosensitivity is again a marked feature, but the condition appears only later in life; porphyrins are produced in excessive quantities, but these are, in general, eliminated in the faeces and do not raise the urinary porphyrin level above normal except during severe transient exacerbations. Photosensitivity is encountered in differing grades of severity, but the crippling deformities of congenital porphyria are rarely seen.

A third disease, acute porphyria, differs in many respects from both congenital porphyria and porphyria cutanea tarda. It is inherited as a dominant character, the highest incidence being in the second or third decade of life, and photosensitivity is never seen. The characteristic symptoms are intense abdominal pain, vomiting, constipation, mental confusion, and, very frequently, progressive paralysis which terminates fatally. In freshly passed urine there is some excess of coproporphyrin, but in place of the uroporphyrin accompanying the other forms of porphyria there is found a colourless, non-fluorescent substance named porphobilinogen, which is transformed on standing into a mixture of uroporphyrins. Porphobilinogen is readily detected, and acute porphyria thus recognized, by treating freshly passed urine with Ehrlich's reagent, a solution of *p*-dimethylaminobenzaldehyde in 5N hydrochloric acid. Figures 11(*a*) and 11(*d*) show respectively urine containing urobilinogen and a urine, from a case of acute porphyria, containing porphobilinogen. A red colour is produced with the reagent if either porphobilinogen or urobilinogen is present; on adding an equal volume of saturated sodium acetate and shaking with chloroform, however, the red compound due to urobilinogen is extracted into the chloroform layer (figure 11(*b*)) while that due to porphobilinogen remains in the aqueous phase (figure 11(*c*)).

The isolation of porphobilinogen has recently been accomplished and it has been shown to be a simple pyrrole derivative (figure 8). On heating in dilute acid it is readily transformed into a mixture of isomeric uroporphyrins in which the series III isomer predominates. This transformation takes place more slowly at room temperature and is hastened by sunlight, so that urine from a case of acute porphyria almost invariably develops uroporphyrin on standing.

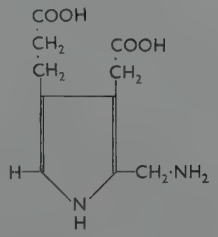


FIGURE 8 – *Porphobilinogen.*

Examination of the organs of a fatal case of acute porphyria shows that porphobilinogen (non-fluorescent) is present in the liver and to a lesser extent in the kidneys and blood; the bone marrow contains neither porphobilinogen nor excessive amounts of porphyrin.

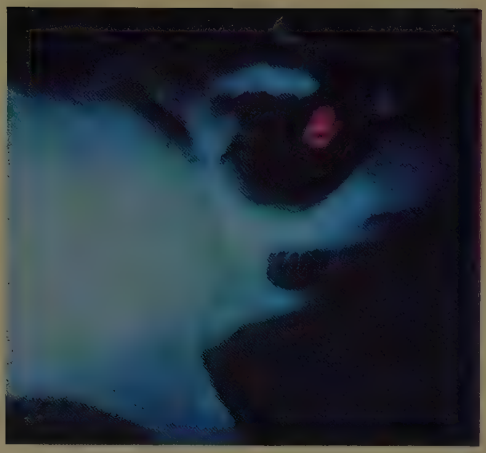


FIGURE 9 – *Rat with gland of Harder exposed. Ultra-violet light.*

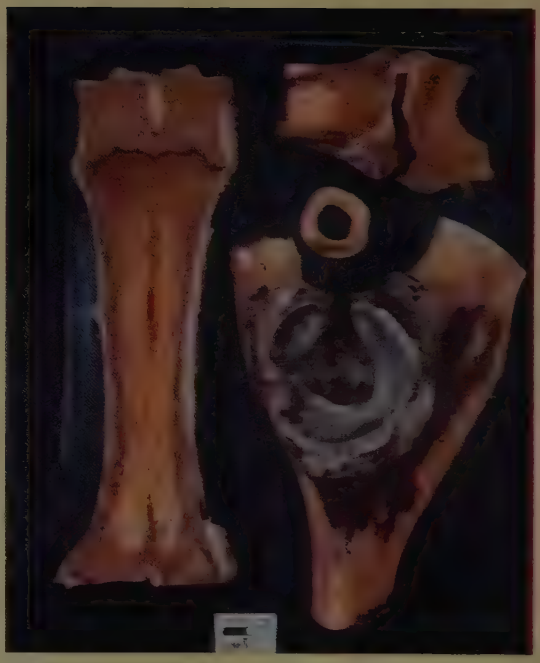


FIGURE 10 – *Congenital porphyria in a South African ox. Bones deeply pigmented with uroporphyrin.*

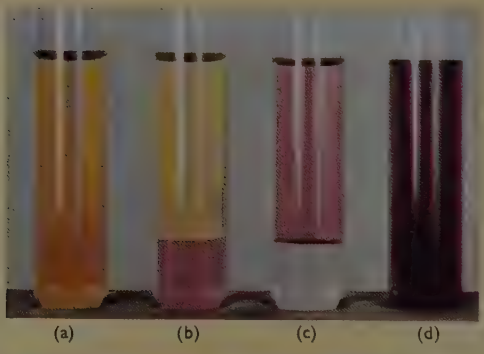


FIGURE 11 (*left*) – *Watson-Schwartz test for porphobilinogen in acute porphyria.*

FIGURE 12 – *Urine containing porphyrin, from a case of congenital porphyria. Ultra-violet light.*

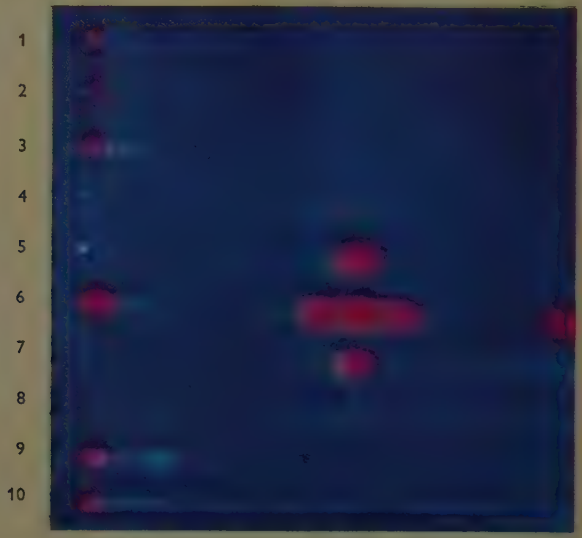


FIGURE 13 – *Paper chromatographic separation of porphyrins. Ultra-violet light.*

FIGURE 14 – *Bones of ground squirrel* (Sciurus niger). (a) *Ultra-violet light.* (b) *White light.*

FIGURE 15 (*right*) – *Tooth of ox with congenital porphyria; portion of rib of a pig with congenital porphyria. White and ultra-violet light. (Specimens lent by Dr T. K. With, Svendborg, Denmark.)*

FIGURE 16 – *Portion of lower jaw of ox with congenital porphyria. Ultra-violet light. (Specimen lent by Dr T. K. With, Svendborg, Denmark.)*

FIGURE 17 – *Upper surface of jaw shown in figure* 16. *Note localized fluorescence due to porphyrin in the teeth. Ultra-violet light.*

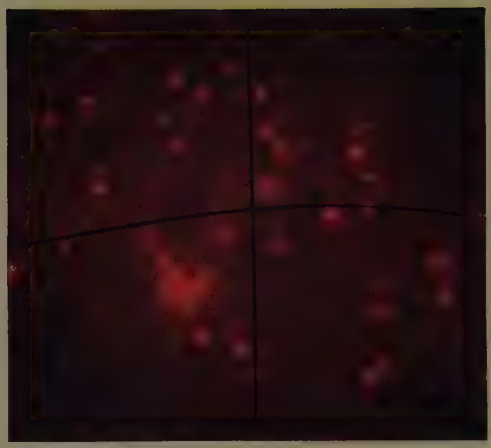


FIGURE 18 – *Bone marrow from a case of congenital (erythropoietic) porphyria in man. Note abundance of cells exhibiting fluorescence due to porphyrin. Ultra-violet light. (Supplied by Professor C. J. Watson and Dr R. Schmid, University of Minnesota.* Arch. intern. Med., **93**, 167, 1954.) (× 260)

FIGURE 19 – *Appearance of blocks of post-mortem liver tissue under ultra-violet light. Left: from cases of congenital (erythropoietic) porphyria. Right: from cases of acute (hepatic) porphyria. Centre: from cases of porphyria cutanea tarda. Note presence of porphyrin only in the latter. (Supplied by Professor C. J. Watson and Dr R. Schmid, University of Minnesota.* Arch. intern. Med., **93**, 167, 1954.)

In congenital porphyria, on the other hand, porphobilinogen is absent and the liver porphyrins are not greatly raised, but the bone marrow contains numerous porphyrin-containing cells of the erythropoietic series which are vividly fluorescent in ultra-violet light (figures 18, 19). In porphyria cutanea tarda the hepatic cells are rich in porphyrin, but contain little or no porphobilinogen. To emphasize the difference in the respective sites of origin of the excessive pigments or their precursors, C. J. Watson has proposed subdivision of porphyria into 'hepatic' and 'erythropoietic' respectively, the former including congenital porphyria and the latter acute porphyria, the cutanea tarda type, and also an experimental porphyria induced by the administration of drugs such as allyl*iso*propylacetylcarbamide, allyl*iso*propylacetamide, and diallylbarbituric acid. All these substances cause the excretion in normal animals of porphobilinogen and of excessive amounts of coproporphyrins in the urine.

THE BIOSYNTHESIS OF PORPHYRINS AND OF HAEM

Ten years ago nothing was known concerning the steps by which the porphyrins and the haem of haemoglobin are synthesized by living cells. Today the principal steps in the biosynthetic sequence have been revealed, a result largely due to the introduction of isotopic labelling techniques. The first notable achievement was the demonstration by D. Shemin and D. Rittenberg [3] in 1946 that the nitrogen atom of glycine is specifically incorporated into the haem of freshly formed erythrocytes in man. Later, duck blood containing nucleated erythrocytes was found capable of carrying out the same biosynthesis *in vitro*; nearly all subsequent work by the American team has employed this system, while British groups have worked with living rabbits or with chicken red-cell preparations.

In all cases the aim has been to degrade the haem, which was synthesized in the presence of an isotopically labelled precursor, in such a way that the degree of labelling of individual atoms can be measured. This is not completely possible in the case of haem, since pyrrole rings I and II are similar in structure and so are rings III and IV; nevertheless, these pairs may be degraded separately. First, the iron is removed from haem and the vinyl groups of the resulting protoporphyrin are catalytically reduced to ethyl groups, the porphyrin formed being known as mesoporphyrin (see figure 20, in which the individual atoms and rings are identified conventionally, A, B, C, and D being used for the latter, however, to avoid confusion with the numerals denoting atoms). Chromic acid oxidation now ruptures the macrocyclic ring, liberating the bridge carbon atoms as carbon dioxide and affording a molecule of

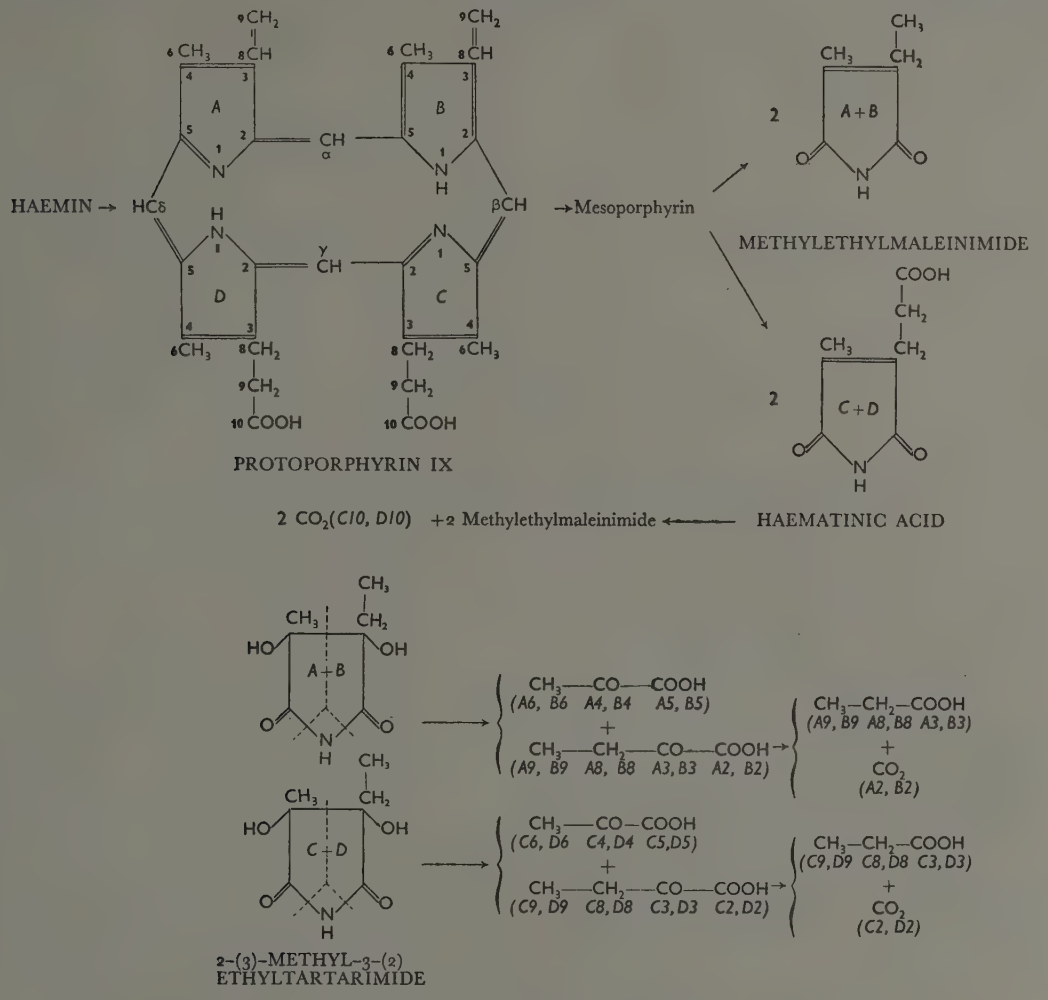


FIGURE 20 – *Degradation of haemin.*

methylethylmaleinimide from each of rings A and B and a molecule of haematinic acid from each of rings C and D. Haematinic acid is decarboxylated by heating to produce a molecule each of carbon dioxide and of methylethylmaleinimide (figure 20). The two lots of methylethylmaleinimide, representing pyrroles A + B and C + D respectively, are separately degraded by the further series of steps shown in figure 20, each carbon atom of the porphyrin thus being finally obtained in the form of carbon dioxide, the radioactivity of which can be measured and compared with that of the ^{14}C substrate used in the experiment.

It has been found that all four nitrogen atoms of haem are derived from the nitrogen of glycine, while the methylene carbon atom of glycine contributes all four bridge carbon atoms and one other carbon atom in each pyrrole ring, namely A_2, B_2, C_2, and D_2. The carboxyl group of glycine is not incorporated into the porphyrin structure. All the carbon atoms of haem not supplied by glycine can be derived from acetate, but experiments with acetate labelled with radioactive carbon in the carboxyl group or methyl group respectively show that acetate itself cannot be the immediate precursor.

If stepwise degradation is applied to the porphyrin synthesized in the presence of acetate labelled with ^{14}C in the carboxyl group, the highest activity is found in the two carboxylic acid groups C_{10} and D_{10} of the porphyrin, although some is present in carbon atoms 3 and 5 of all

rings. When acetate labelled with ^{14}C in the methyl group is used, the highest activity is found in carbon atoms 6 and 9 of all rings but a great deal is found also in positions 4, 8, 5, and 3 and a little in C_{10} and D_{10} (see table I). Evidence that the two halves of the pyrrole rings which give rise to pyruvic acid and α-ketobutyric acid respectively on degradation (figure 20) are derived from the same precursor is forthcoming from equal labelling, each to each, of the position pairs 6 and 9, 4 and 8, 5 and 3 respectively (figure 21).

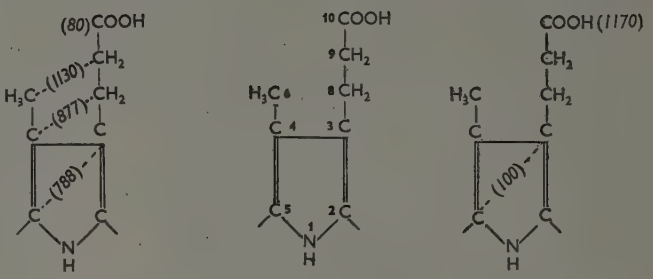


FIGURE 21 – *Average activities, in parentheses, of comparable carbon atoms in all pyrrole units. The pyrrole unit represented contains a carboxyl group, found only in rings C and D of the porphyrin. (After Shemin and Wittenberg* [4].)

Shemin and Wittenberg [4] postulated from these findings that in the course of porphyrin biosynthesis a single precursor of a pyrrolic nature was first formed from union of two unsymmetrical 4-carbon units and one glycine unit. The 4-carbon unit could be a derivative of succinic acid arising from acetate *via* the tricarboxylic acid cycle. If this were so, the precursor of all four rings A to D would carry acetic acid and propionic acid side chains in its ββ′ positions. Union in some way of four units of this pyrrole and four units representing the α carbon atoms of glycine molecules could yield a uroporphyrin. Postulated loss of six carboxyl groups and four hydrogen atoms could then lead to protoporphyrin and so, after the incorporation of iron, to haem. The scheme is summarized below (compiled after Shemin):

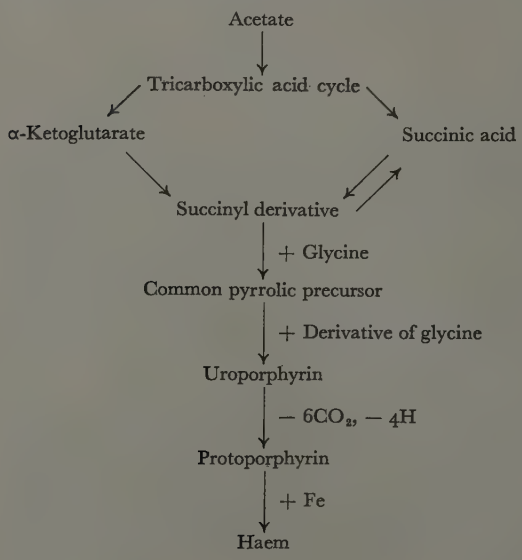


TABLE I

Total activity (counts per minute) of each of the carbon atoms of protoporphyrin biosynthesized from acetate labelled with ^{14}C in the methyl and carboxyl group respectively

Carbon number	[*methyl-*^{14}C] *acetate*		[*Carboxyl-*^{14}C] *acetate*	
	Rings A + B	*Rings C + D*	*Rings A + B*	*Rings C + D*
2	0	0	0	0
3	776	767	95	88
4	973	811	0	0
5	770	833	109	106
6	1210	1010	0	0
8	854	869	0	0
9	1190	1130	0	0
10	—	80	—	1260

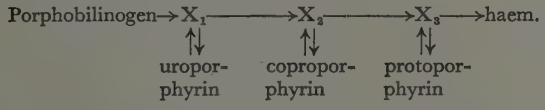
The fact that the addition of coenzyme A to an avian blood system increases the utilization of succinate for porphyrin formation *in vitro* suggests that the unsymmetrical succinyl derivative may be succinyl coenzyme A, but no direct evidence in support of this is as yet forthcoming.

When pure porphobilinogen became available and G. H. Cookson and C. Rimington [5] had shown that it was a pyrrole derivative (figure 8) carrying the same side chains in the β and β′ positions as are present in the pyrrole rings of uroporphyrin (figure 7), its possible relation to normal porphyrin biosynthesis became obvious.

It was shown by J. E. Falk, E. I. B. Dresel, and C. Rimington [6] that porphobilinogen added to an avian red-cell haemolysate could serve as a substrate for porphyrin production. Proof that it was, in fact, the 'common pyrrolic precursor' of the above scheme and a true intermediate in normal porphyrin biosynthesis, followed from the

further demonstration that radioactive porphobilinogen could be isolated from a haemolysate utilizing radioactive glycine as substrate. Since glycine and succinic acid had already been established as raw materials for the synthesis of porphyrins, it became clear that some structure derived from their union must intervene between them and porphobilinogen in the train of chemical events. Shemin and C. S. Russell [7] in America, and almost simultaneously A. Neuberger and J. J. Scott [8] in England, identified this structure as δ-aminolaevulic acid. According to Shemin, glycine and 'active' succinate first combine to form α-amino-β-ketoadipic acid, which, being a β-ketonic acid, readily loses carbon dioxide (hence the absence of the carboxyl carbon of glycine from the final porphyrin structure) to yield δ-aminolaevulic acid. Neuberger's group have purified the enzyme which condenses two molecules of δ-aminolaevulic acid and produces porphobilinogen. The process is shown in figure 22.

So far, little has been said concerning the position of uroporphyrin, coproporphyrin, and protoporphyrin in the biosynthetic pathway leading to haem. They would appear to be logical steps, but the evidence is rather conflicting. Certainly red-cell haemolysate systems can bring about some porphyrin interconversions, but they do so rather inefficiently, and radioactive labelling data show that much better biosynthesis results when either glycine and succinate, δ-aminolaevulic acid, or porphobilinogen acts as substrate. Considerations of the permeability of the sub-cellular particles may be responsible for this difference. Alternatively, it is possible that the porphyrins are not on the direct line of biosynthesis but are produced by reversible side reactions from closely related precursors, possibly the more highly reduced porphyrinogens, as follows:



The exact nature of the porphyrins produced in these systems and the isomeric series to which each belongs is at present being very carefully investigated by paper chromatographic and other techniques. Figure 13 illustrates the use of paper chromatography for this purpose. (1–3) are uroporphyrins from *in vitro* biosyntheses; (4–5) coproporphyrin III from a similar source; (6) markers, respectively uroporphyrin I (on left), coproporphyrins I, III, and II, and deuteroporphyrin IX; (7) coproporphyrin III produced by decarboxylating uroporphyrin; (8–10) uroporphyrins from *in vitro* biosyntheses. Before long, the entire chain of events by which the body synthesizes its vital haem and porphyrin pigments should become known, and possibly also the nature of the chemical aberrations which are responsible for porphyria diseases of man and animals.

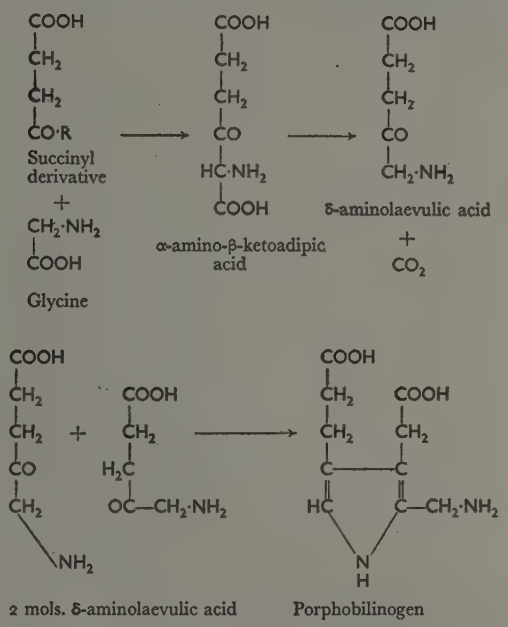


FIGURE 22 – *Scheme summarizing biosynthetic steps from glycine + succinate (from the tricarboxylic acid cycle) to porphobilinogen, the simplest pyrrolic substance known to be a precursor of haem and porphyrins.*

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George Johnston and the Ray Society

C. M. YONGE

Natural history societies have long made valuable contributions to biology, and in this year fall two important anniversaries in their history. It is the 250th anniversary of the death of John Ray, among the greatest of British naturalists, and the 100th anniversary of the death of George Johnston, founder of the Ray Society and a noteworthy representative of the early nineteenth-century naturalists who did so much to establish the identity and distribution of British species of plants and animals. The Ray Society is internationally known for its beautifully illustrated volumes dealing with British natural history.

Linnaeus inspired a fashionable interest in natural history in the eighteenth century. His method of description and of naming, and his system of classification—however artificial this later proved to be—appealed to the orderly minds of that age of reason. A further stimulus came from the voyages in the Pacific and Indian Oceans of Cook and other explorers who brought home a wealth of new plants and animals. These were all named and classified, and then found their way into national museums or into the cabinets of wealthy aristocratic collectors.

Apart from Linnaeus, the leading naturalist in Europe at that time was George Louis Leclerc, Comte de Buffon. No gentleman's library was complete without his magnificently illustrated *Histoire naturelle*. In Britain Sir Joseph Banks guided the development of natural history with as much authority as he did that of the Royal Society. The first 'British Zoology' was produced, in the intervals between his numerous tours, by Thomas Pennant, a country gentleman of liberal means. His friend and correspondent, Gilbert White, with little money but with leisure and cultured interest, built a more enduring monument within the narrow confines of Selborne.

We know less of John Ellis, who revealed the purely animal nature of zoophytes and helped to describe the corals collected by Cook, but he was of sufficient consequence to be asked to display his collections to the Princess of Wales. Colonel George Montagu, a somewhat later figure, was a gentleman of wealth who retired from the army to study the animals of Britain's western shores and to write books on birds and molluscs. The few who made a living, as opposed to a hobby, out of natural history did so by catering to the taste for illustration. Edward Donovan serves as an example; his books on birds, fishes, molluscs, and insects have little beyond the beauty of their plates to commend them.

As the nineteenth century advanced the influence of the landed gentry began to decline, while that of the middle classes rose. The wider spread of education and of some measure of wealth and leisure were reflected in a wider interest in natural history and, in particular, in the plants and animals of the countryside, shore, and sea. The ranks of the amateur naturalists were reinforced by professional men, in particular doctors and clergymen, and by small merchants and shopkeepers. Later in the century came the self-taught artisans like William Peach, Hugh Miller, and Thomas Edward. Gradually there emerged the professional zoologist and botanist, although for much of the nineteenth century material reward was as meagre as enthusiasm and achievement were great.

Of these new naturalists, George Johnston stands out as a truly representative figure well worthy of remembrance. The tenth of a family of fifteen, he was born on a farm at Simprin in Berwickshire, on 27th July, 1797. He was educated at Berwick before proceeding to the University of Edinburgh to study medicine. When twenty years old he obtained the diploma of the College of Surgeons, and after some further study in London, and a brief period of practice in Northumberland, he settled permanently in Berwick-on-Tweed. He built up the largest medical practice in Berwick and the surrounding countryside, and became a familiar and much-loved figure.

He devoted every available moment to the study of natural history, on which he wrote many papers and several books. He conducted a voluminous correspondence with contemporary naturalists and much of this has, most fortunately, been preserved and published. A book of 'Selections from the Correspondence of Dr George Johnston,'

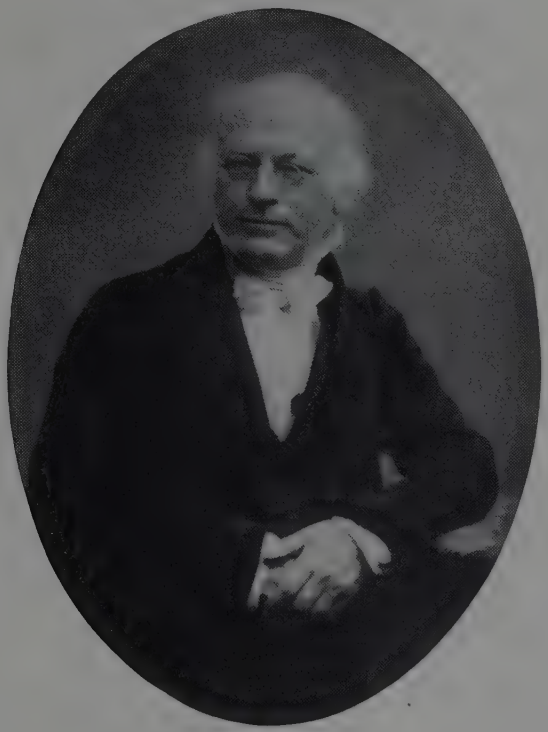


FIGURE 1 – *Dr George Johnston, from a photograph taken in later life.*

collected and arranged by his daughter, Mrs Barwell-Carter, appeared in 1892, to celebrate the sixtieth anniversary of the foundation of the Berwickshire Naturalists' Club.

These letters form a volume of over 500 pages, and are addressed largely to naturalists. They include P. J. Selby, the ornithologist; Joshua Alder of Newcastle, who was part-author of 'The British Nudibranchiate Mollusca'; William Thompson, of Belfast, who died too early to produce more than the early volumes of a projected natural history of Ireland; the Reverend David Landsborough of Saltcoats, Ayrshire, the author of 'Excursions to Arran' and various books on marine biology; Charles William Peach, at that time in the customs office at Fowey; and also local naturalists such as James Hardy and Archibald Hepburn. Interspersed with these are letters to his publishers, W. H. Lizard of Edinburgh and John Van Voorst of London, while in later years many are addressed to Mrs Alfred Gatty, author of 'Parables of Nature' and of many books for children, who shared his interest in the natural history of seaweeds.

Almost every contemporary British naturalist is mentioned in these letters. Although Mrs Barwell-Carter arranged the letters, they were edited by James Hardy, at that time honorary secretary of the club, who greatly increased the value of the book by his informative footnotes dealing with almost every person mentioned. Letters and notes together form a mine of information about British naturalists and their activities between 1830 and 1853.

The Berwickshire Naturalists' Club, the first provincial society of its kind in Britain, was founded in 1831 'for the purpose of examining the Natural History and Antiquities of the county and its adjacent districts and of affording to such as were interested in these objects, the opportunity of benefiting by mutual aid and co-operation.' There were initially eleven members, and Johnston was the founder and first president. The letters do not deal with the foundation of the club, but its later activities are frequently mentioned. Johnston had clear views about the functions of such societies. Writing to Joshua Alder, he declines the offer of a mere catalogue of the names of local molluscs, 'believing as I do that it is the prime business or duty of provincial societies to furnish their members with the means of investigating the natural history of their own district without sending them to other works or museums.'

FIGURE 2 – *Long-tailed shark. An illustration from Johnston's description of a specimen captured at Berwick in* 1846, *and published in the 'History of the Berwickshire Naturalists' Club.'*

FIGURE 3 – *Sea anemone,* Bolocera tuediae *(Johnston), described (as* Anthea tuediae*) in 'A History of the British Zoophytes.'*

FIGURE 4 – *Sponge growing on the back of a spider crab, figured in 'A History of British Sponges and Lithophytes.'*

In 1834 he sends a full account of a meeting to a member who had been absent. 'A paper by William Baird on the Berwickshire Entomostraca was read. He knows already more Berwickshire species of these very curious creatures than have been yet enumerated as British, and the paper, when completed, promises to be a most interesting one. . . . I then exhibited some Birds, Fishes, and Insects, which had been taken lately in Berwickshire. . . . The Grey Phalarope was among the birds. Embleton again brought the Crabs and Lobsters of the coast before us, naming them from my collection, in which there were six additional species to his list, and a fine species of *Galathea,* which I think is new. . . . I now exhibited a collection of the Bivalved Shells of the coast, and read a paper on them. . . .'

The letters, with their descriptions of such meetings, and of inquiries and discussions about specimens of all kinds, show clearly that Johnston and his contemporaries were working their way through the local fauna and flora, puzzling out the identity of animals and plants that are now familiar. All the time they were adding new species to their local lists and so building up that knowledge about the nature and distribution of British species which was the particular contribution of these early nineteenth-century naturalists.

There is no concern with biological evolution. The 'Origin of Species' was not published until four years after Johnston died, and he had clearly no thought but that all forms of life originated from a beneficent and endlessly ingenious Creator. He was a describer of species and an observer of habit and distribution, but he was no comparative anatomist. On several occasions, when he is tempted to apply for academic posts, this ignorance of internal structure is realized to be a drawback. He admits that he 'was educated in an earlier school, and am not likely ever to rise above, if I could even equal, my masters: these were Pennant and Montagu, who have merit certainly, but they were the founders of the school of superficiality.' Certainly no major discovery came from Johnston, but he was receptive to those of others. After going to much trouble to confirm

them, he accepts Grant's description of the water currents in sponges and the discovery by Vaughan Thompson, revolutionary in its day, that barnacles are not molluscs but crustaceans.

Johnston wrote five books. An early 'Flora of Berwick' (1829–31); 'A History of British Zoophytes' (first edition, 1838; second edition, 1847); 'A History of British Sponges and Lithophytes' (1842); 'An Introduction to Conchology' (1850); and finally 'The Natural History of the Eastern Borders,' of which only the first volume, dealing with botany, appeared (1853). None are works of the first importance, although the book on zoophytes was the first published upon them since that of John Ellis in 1755. It represents a stepping stone between that book and the later volumes of Gosse, Allman, Hincks, and, most recently, Stephenson. Today, the most readable section is the chapter on the history of 'zoophytology.' The book on conchology was praised by Edward Forbes and was even translated into German, but it is no more than an interesting compilation about shells and their inhabitants. 'The Natural History of the Eastern Borders' was to have been his crowning work, and it is sad that only one volume was completed; this is full of interesting local information. The club published, and still publishes, its 'History' which he edited, and he was also part-editor of a short-lived 'Magazine of Zoology and Botany'; this merged into the 'Annals and Magazine of Natural History,' which continues to appear.

There is little doubt, however, that Johnston's name will always be remembered primarily in connection with the foundation of the Ray Society. First mention of this in the 'Correspondence' is in a letter to James Hardy, dated 20th February, 1843. But he is soon writing to all his friends on the subject, begging them to help by interesting others. After discussing the difficulties of publishing books on natural history at a reasonable price, he writes: 'I have long wished to propose to establish a Ray Society, at an annual subscription of one or two sovereigns, to print works illustrative of the Natural History of Great Britain, original and old.' And later, in an almost impassioned letter to William Thompson, he writes that with the establishment of this society 'I shall see the neglected corners of our Fauna and of our Flora searched out. . . . Natural History is a good study and a wholesome one, and the meat and medicine have been too long dispensed only to the rich. . . . It is because I am poor that I plead for the Ray Club.'

In the obituary of Sir William Jardine, published in the 'History' of the Berwickshire Naturalists' Club, of which he was at one time president, some credit for the foundation of the Ray Society is given to Hugh Strickland, his son-in-law. The latter certainly assisted Johnston by distributing circulars and collecting members. It was hoped to secure five hundred members, but after initial optimism Johnston had a period of despair. However, there was little reason for this, because by April 1844 he was able to report that the Ray Society had made a beginning. By September of the same year it had assumed 'a more healthy appearance. It has increased considerably of late, and we have one large volume in the press, which will be ready about Christmas.'

In point of fact, such was the interest that his enthusiasm inspired that considerably over five hundred members were enrolled within the first year. Johnston had done a major and an enduring service to the cause of natural history. During the 111 years that have elapsed since its foundation, the Ray Society has published some 140 volumes, dealing with every aspect of British natural history —from protozoans to whales, from lichens to mites and barnacles. Many of these monographs rank high among similar works in any language. The society's highest membership, 868, was attained within five years of its foundation, after which numbers slowly declined until they fell to less than two hundred in the years between the two world wars. There was a real danger that the society would have to be wound up during the last war, but this was fortunately averted owing to generous donations, particularly from Imperial Chemical Industries Limited. Membership has since increased, and it is now well over four hundred. Johnston would view with much happiness the hopeful prospects, in its second century, of the society he did so much to establish.

After settling down in practice in Berwick Johnston rarely left home. Writing in 1853, he says that 'during the thirty-five years I have been in Berwick I have not had six weeks' holiday in all, rather, I should say it has been one grand holiday; . . . ' The esteem in which he was held is shown by the long period for which he served as Alderman and by the fact that he was thrice elected Mayor. These activities he certainly enjoyed, but probably most of all he loved the meetings of the club he had founded and their excursions—perhaps to Holy Island with its prospect of 'the priory, and church, and castle, and Bamborough Castle, and the Farne Islands, and the sea, the glorious sea, . . .' He died in 1855.

Organic compounds of the metals

W. WARDLAW and D. C. BRADLEY

The chemistry of organometallic compounds presents many points of practical as well as theoretical significance. Within the last few years the silicones, for example, which less than twenty years ago seemed to be of only academic interest, have become of very considerable industrial importance. Organic compounds are likely to prove useful for the purification of some of the rarer elements, such as zirconium, which have lately become of interest to engineers.

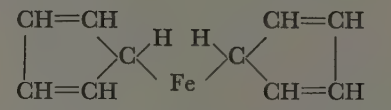
Organic derivatives of metals and metalloids have long been known, and their study has led to many important developments in both pure and applied chemistry. Compounds containing carbon directly linked to a metallic element are now often termed organometallic, to distinguish them from the wider class of organic compounds of the metals which need not contain metal-carbon bonds.

The great importance of organomagnesium compounds was first recognized by Grignard in 1900. An unusual use of the Grignard reagent is in the preparation of the aromatic hydrocarbon diphenyl, made by the action of phenyl magnesium bromide on anhydrous ferric chloride.

$$6C_6H_5MgBr + 2FeCl_3 = 2Fe + 3MgBr_2 + 3MgCl_2 + 3C_{12}H_{10}\ (\text{diphenyl}).$$

T. J. Kealy and P. L. Pauson [1] used this method in an attempt to prepare the hydrocarbon fulvalene ($C_{10}H_8$), and discovered a remarkable new compound which contained only carbon, hydrogen, and iron. By reaction between ferric chloride and cyclopentadienyl magnesium bromide (C_5H_5MgBr) they hoped to obtain dicyclopentadienyl ($C_{10}H_{10}$) as an intermediate step to their goal. They isolated instead an orange, ether-soluble compound of iron, having m.p. 172·5–173° C and the formula $Fe(C_5H_5)_2$. It had great chemical stability and concentrated nitric acid was required to decompose it. Further work [2] revealed that it volatilized without decomposition into a monomeric, undissociated vapour which obeyed the perfect-gas laws even at 400° C.

G. Wilkinson, M. Rosenblum, M. C. Whiting, and R. B. Woodward [4] found that the compound was diamagnetic and without appreciable dipole moment. In addition they examined its optical absorption spectrum and concluded that the substance contained only one type of C—H bond, in opposition to the structure (I) proposed by Kealy and Pauson.



(I)

They suggested the sandwich structure shown in figure 1, and X-ray studies by three groups of workers [5] have confirmed their views.

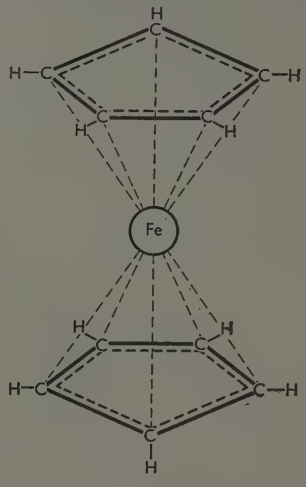


FIGURE 1

Further research [4] disclosed that the compound was readily oxidized in solution to a bright blue cation $[Fe(C_5H_5)_2]^+$, which was isolated as the crystalline tetrachlorogallate.

Woodward, Rosenblum, and Whiting [6] discovered that the compound would participate in the Friedel-Crafts reaction and that the ring hydrogen was substituted without affecting the metal atom.

$$Fe(C_5H_5)_2 + 2CH_3COCl \xrightarrow{Al_2Cl_6} Fe(C_5H_4.COCH_3)_2 + 2HCl.$$

This elegant demonstration of the aromatic character of iron biscyclopentadienyl led to the suggestion that the compound should be named

ferrocene. Attention was then directed to the preparation of similar derivatives of other transition metals. In the space of two years cyclopentadienyl derivatives of most of the transition metals have been made by Wilkinson and his collaborators at Harvard University, and by E. O. Fischer and his collaborators at Munich. Some examples of these compounds are given in table I.

In addition to the above compounds, some analogous compounds of indene have been prepared, such as $Fe(C_9H_7)_2$, $[Co(C_9H_7)_2]^+$, and $Ni(C_9H_7)_2$. The table shows that the formation of stable biscyclopentadienyl derivatives seems to be a typical property of the transition metals.

In the structure established for the ferrocene molecule the interatomic distance for the carbon atoms in the rings is 1·4 A, in accord with aromatic substances generally. For ferrocene and the other biscyclopentadienyl metal derivatives there is a characteristic infra-red absorption at 3·27 μ corresponding to a single C—H bond-stretching, which is typical of C—H bonds in aromatic systems. This suggests that all compounds such as those shown in table I have similar structures. Magnetic measurements [8] have yielded valuable information about the electronic arrangements in these new substances. Many, such as $Fe(C_5H_5)_2$ and $[Co(C_5H_5)_2]^+$, are diamagnetic, but others, for example $[Fe(C_5H_5)_2]^+$ and $Co(C_5H_5)_2$, are paramagnetic, consistent with the presence of a single unpaired electron. $[V(C_5H_5)_2]^+$, $Cr(C_5H_5)_2$, and $Ni(C_5H_5)_2$ are paramagnetic and the results indicate the existence of two unpaired electrons in each ion or molecule.

Measurements of the heats of combustion of ferrocene and of biscyclopentadienyl nickel [9] indicate that the nickel compound has the smaller thermodynamic stability.

Wilkinson and J. M. Birmingham [10] have shown that when present in these metal-organic systems the metals of Groups IVA and VA still retain 'transitional' properties.

A common property of the biscyclopentadienyl cations is the formation of insoluble salts with large anions such as picrate, ferrocyanide, and silicotungstate.

The methods developed for the preparation of the biscyclopentadienyl compounds may be considered under four headings:

1. Reactions of metal halides or acetylacetonates with either cyclopentadienyl magnesium halide or cyclopentadienyl sodium. This method has been extensively used by Wilkinson and his collaborators, e.g:

$$TiCl_4 + 2NaC_5H_5 \rightarrow Ti(C_5H_5)_2Cl_2 + 2NaCl.$$

2. Reaction of a complex metal ammine with either lithium, sodium, or potassium cyclopentadienyls in liquid ammonia, followed by thermal dissociation of the metal ammine cyclopentadienyl. This method was devised by Fischer and his collaborators [11], e.g:

$$Co(NH_3)_4(SCN)_2 + 2KC_5H_5 \xrightarrow{\text{liq. } NH_3} Co(NH_3)_4(C_5H_5)_2 + 2KSCN.$$

$$Co(NH_3)_4(C_5H_5)_2 \xrightarrow{\text{Heat}} Co(C_5H_5)_2 + 4NH_3.$$

3. Reaction of iron and cyclopentadiene [3]:

$$Fe + 2C_5H_6 \xrightarrow{375^\circ C} Fe(C_5H_5)_2 + H_2.$$

4. Wilkinson [7, 9] has reported a method which involves vapour phase reaction between a metal carbonyl and cyclopentadiene. The carbonyls of chromium, iron, cobalt, and nickel can all be converted to the neutral biscyclopentadienyls, e.g. $Cr(CO)_6$ to $Cr(C_5H_5)_2$. In the cases of molybdenum or tungsten

TABLE I

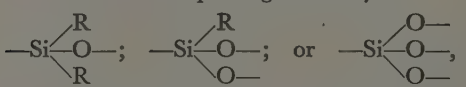
Group				
IVA	*VA*	*VIA*	*VIIA*	*VIIIA*
$Ti(C_5H_5)_2$	$V(C_5H_5)_2^+$	$Cr(C_5H_5)_2$	$Mn(C_5H_5)_2$	$Fe(C_5H_5)_2$
$Ti(C_5H_5)_2^+$	$V(C_5H_5)_2X_2$ (X = Cl or Br)	$C_5H_5Mo(CO)_5MoC_5H_5$		$Fe(C_5H_5)_2^+$
				$Ru(C_5H_5)_2$
$Ti(C_5H_5)_2X_2$ (X = F, Cl, Br, or I)	$Nb(C_5H_5)_2Br_3$	$[(C_5H_5)_2Mo^{IV}Cl][Cr(CNS)_4(NH_3)_2]H_2O$		$Ru(C_5H_5)_2^+$
				$Co(C_5H_5)_2$
$Ti(C_5H_5)_2(OH)Br$, H_2O	$Nb(C_5H_5)_2(OH)Br_2$	$Mo(C_5H_5)_2Cl_2$		$Co(C_5H_5)_2^+$
$Zr(C_5H_5)_2Br_2$	$Ta(C_5H_5)_2Br_3$	$[(C_5H_5)_2Mo^{V}Cl_2]_2[PtCl_6]$		$Rh(C_5H_5)_2^+$
		$[(C_5H_5)_2W^{V}Cl_2]_2[PtCl_6]$		$Ir(C_5H_5)_2^+$
				$Ni(C_5H_5)_2$
				$Ni(C_5H_5)_2^+$

carbonyls, the remarkable cyclopentadienyl carbonyls were isolated, e.g.

$$2Mo(CO)_6 + 2C_5H_6 \rightarrow C_5H_5Mo(CO)_5MoC_5H_5 + CO, \text{ etc.}$$

Earlier discoveries in the field of organometallic chemistry posed problems in valency theory and this has certainly been true with respect to the metal biscyclopentadienyls. The electronic configurations of these compounds has clearly revealed the limitations of current theories, but this fact will no doubt act as a stimulus [8, 12].

The new compounds which have just been discussed are as yet of academic interest only. However, the organic silicon compounds to which Kipping, at Nottingham, devoted his attention from 1899 to 1939 are now of wide industrial application. At no time did Kipping consider that the materials he had made would have any commercial value. His view was expressed in 1937 in the Bakerian Lecture to The Royal Society [13] when he said 'the prospect of any immediate and important advance in this section of chemistry does not seem to be very hopeful.' Yet by the late thirties American interests were examining the possibilities of commercial production of some of the products regarded by Kipping as of little value. A company to manufacture organic silicon compounds was formed in 1943. Recently a new industry based on these remarkable chemical compounds has been started in Britain. The substances concerned are the silicones. The name was used originally to describe compounds of the empirical formula $R_1R_2Si{=}O$, since these were expected to be analogous to the ketones $R_1R_2C{=}O$. This empirical formula can denote an infinite number of compounds, and the term silicone is now generally applied to all organo-silicon compounds having at least one Si—O—Si group. The silicones consist of chain, ring, or cross-linked chain polymers in which the repeating unit may be



R being an alkyl or aryl group. Methyl silicones are the most important (for detailed information consult references [14] and [15]). The mode of formation of the silicones can best be understood by considering the mechanisms of hydrolysis of the three key types of organosilicon compound: R_3SiX, R_2SiX_2, and $RSiX_3$, where R is an alkyl or aryl group and X is chlorine or ethoxy.

Trimethyl-chlorosilane $(CH_3)_3SiCl$ is readily hydrolysed to the unstable silanol, $(CH_3)_3SiOH$, which itself condenses to form a stable compound, $(CH_3)_3Si{-}O{-}Si(CH_3)_3$, which boils at 99·5° C:

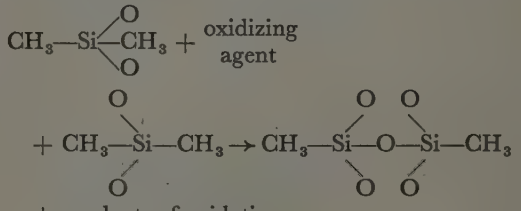
$$(CH_3)_3SiCl + H_2O \rightarrow (CH_3)_2SiOH + HCl$$
$$2(CH_3)_3SiOH \rightarrow (CH_3)_3Si{-}O{-}Si(CH_3)_3 + H_2O.$$

Triaryl chlorosilanes can be converted into triaryl silanols. These compounds, e.g. $(C_6H_5)_3SiOH$ (m.p. 150–151·5° C), are relatively stable and easily isolated. The stability of the carbon-silicon bonds in organosilicon compounds and of the siloxane grouping Si—O—Si accounts for the stability of the silicones.

The hydrolysis of $(CH_3)_2SiCl_2$ leads to the formation of the very unstable dimethyl-silanediol $(CH_3)_2Si(OH)_2$. Again, the corresponding aryl derivatives, such as $(C_6H_5)_2Si(OH)_2$ (m.p. 148° C), are much more stable than the alkyl compounds. The following equation illustrates the type of condensation which may occur:

$$2(CH_3)_2Si(OH)_2 \rightarrow HO.(CH_3)_2Si{-}O{-}Si(CH_3)_2OH + H_2O$$

The hydrolysis of $RSiX_3$ will obviously lead to complex products and, in general, hard brittle solids are ultimately obtained. By varying the proportions of reactive intermediates and the conditions of hydrolysis, however, a wide range of silicones may be obtained; these include mobile non-volatile liquids, highly viscous greases, and hard brittle resins. The products can be further modified by chemical treatment. For example, by the action of ferric or aluminium chloride the linear polymers are converted into rubberlike gels which will undergo cross-linking when subjected to oxidation by organic peroxides. It is believed that the oxidation process involves the elimination of methyl groups, allowing cross-linkage to occur:



+ products of oxidation.

The outstanding development in silicon chemistry in recent years is the discovery by W. I. Patnode and E. G. Rochow [16] in 1945 of a method of preparing organosilicon intermediates directly from silicon and alkyl halides. In this direct synthesis the vapour of alkyl or aryl halide is passed over a heated, finely divided mixture of silicon with either copper or silver. Various alkyl or aryl chlorosilanes are formed. When methyl

chloride is used with a silicon-copper mixture in the temperature range 250–280° C, the products consist chiefly of CH_3SiCl_3 (b.p. 66° C) and $(CH_3)_2SiCl_2$ (b.p. 70° C), together with some $(CH_3)_3SiCl$ (b.p. 57·6° C) and CH_3SiHCl_2 (b.p. 42° C). The separation of these products presents a difficult problem, for their boiling points lie close together; in addition the substances are inflammable, toxic, and corrosive.

In general, it is the unusual physical properties of silicones which have determined their special uses. The dimethylsiloxane polymers,

$$(CH_3)_3Si—O—[Si(CH_3)_2—O]_n—Si(CH_3)_3,$$

are colourless fluids ranging from mobile volatile lower members to non-volatile highly viscous long-chain compounds. Silicone rubber far surpasses other elastic materials in its resistance to heat and oxidation, and its elasticity is maintained over the remarkably wide temperature range — 90° C to 250° C. Nevertheless, the chemicals that swell ordinary rubber also swell silicones. The most striking property of a silicone fluid is its very low temperature-coefficient of viscosity. This behaviour suggests that intermolecular forces in silicones are abnormally weak—a view that is confirmed by the low surface tensions and high compressibilities of silicones. In addition, silicones are strongly water repellent, and this property, combined with admirable dielectric behaviour and high thermal stability, has established a place for silicones in the electrical industry. For the dimethyl silicone polymers the dielectric constant approaches a limiting value of c 2·82 as the length of the polymer chain increases ($n > 500$). These fluids have high dielectric strengths (c 100–120 kV/cm at 0·254 mm to 200 kV/cm at 2·54 mm). The volume resistivity is high (c 10^{14} ohm.cm) and remains sensibly constant up to 200° C. In addition, power factors are low and not appreciably affected by temperature or frequency of applied electric field. Thus power factors vary from 0·0001 at 10^3 c/s to 0·0006 at 10^8 c/s. The performance of glass and ceramic insulators is improved by a thin coating of silicone fluid, which prevents the formation of a coherent layer of water under humid conditions. Silicone fluids prevent surfaces of various materials from sticking and they are used extensively as mould-release agents in metal die-casting. The ability of films of silicones to repel water has been utilised in the waterproofing of clothing, and in making furniture and motor-car polishes.

Silicone fluids are essentially linear polymers, but by the introduction of a certain degree of cross-linking silicone greases are obtained. Research workers are finding that silicone greases are invaluable for lubricating stopcocks or ground glass joints in high vacuum systems (10^{-6} mm. Hg). The silicone resins resist oxidation, ultra-violet light, and chemical fumes. Masonry, concrete, and brickwork are made more durable by treatment with them.

The discovery of the remarkable properties of the silicones has stimulated research into the organic compounds of related elements. Germanium, lead, and tin form alkyl and aryl derivatives, but no products analogous to the silicones have been reported. Recently the preparation of unstable alkyl and aryl compounds of titanium was announced [17].

So far in this article, attention has been directed to organic derivatives of the elements in which the element is directly linked to the carbon. Mention should, however, be made of an important group of substances known as the orthoesters or alkoxides, in which the link with the central atom is through the oxygen atom in the OR radical, where R is methyl, ethyl, etc. Such substances as the silicon esters and titanium esters, $Si(OR)_4$ and $Ti(OR)_4$, find wide use in industry and have a fascinating chemistry. In recent years a detailed study has been made of a wide range of the metal alkoxides and the results are of unusual interest [18].

Although silicon and titanium are members of the same family of chemical elements they show marked differences in their own reactions and in the reactions of their compounds. The reaction between silicon tetrachloride and alcohol proceeds to completion in accordance with the equation

$$SiCl_4 + 4EtOH \rightarrow Si(OEt)_4 + 4HCl$$

Titanium tetrachloride, on the other hand, is converted under the same conditions to

$$Ti(OEt)_2Cl_2.EtOH,$$

which is stable even on prolonged boiling with alcohol. Preparation of the orthoester involves reaction between titanium tetrachloride and alcohol in the presence of anhydrous ammonia.

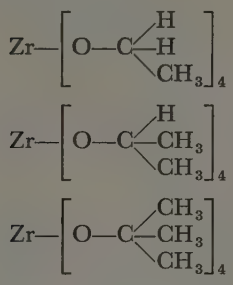
$$TiCl_4 + 4ROH + 4NH_3 = Ti(OR)_4 + 4NH_4Cl.$$

Butyl orthotitanate is made commercially by this method, and confers upon paints excellent heat-resisting properties. Zirconium shows important relationships to both silicon and titanium, yet it is distinctive in being capable of exhibiting a covalency of eight: the maximum covalency exhibited by silicon and titanium is six. The high

covalency of zirconium is reflected in the range and complexity of its compounds. Zirconium ethoxide is formed when alcohol and zirconium tetrachloride react in the presence of ammonia.

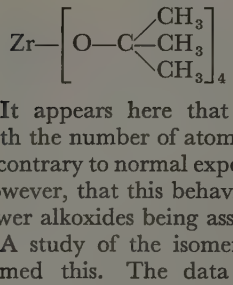
$$ZrCl_4 + 4ROH + 4NH_3 \rightarrow Zr(OR)_4 + 4NH_4Cl.$$

An outstanding feature of these zirconium alkoxides is the increase in volatility as the alkyl group is changed from primary to secondary to tertiary. Thus:



b.p. 180° C (0·1 mm)

b.p. 160° C (0·1 mm)



b.p. 50° C (0·1 mm)

It appears here that the volatility increases with the number of atoms in the molecule, which is contrary to normal experience. It can be shown, however, that this behaviour is in fact due to the lower alkoxides being associated.

A study of the isomeric amyloxides has confirmed this. The data for titanium and zirconium amyloxides are presented in table II where M = molecular weight (determined ebullioscopically in benzene) and X = molecular complexity or degree of polymerization (i.e. M/(formula weight)).

It is evident from table II that the molecular complexity and the boiling point of a zirconium amyloxide is determined by the degree of branching of the amyl groups. The same effect is apparent, but to a lesser extent, with the amyloxides of titanium. These data are best interpreted on a stereochemical basis. Thus the straight chain alkoxides tend to polymerize by intermolecular bonding between the central atom and the oxygen atoms from alkoxide groups. This is a consequence of the well known covalency expansion which is exhibited by titanium and zirconium. Figure 2 illustrates a possible structure for a trimeric alkoxide. It is easy to see that the rotation of highly branched alkoxide groups will preclude the formation of intermolecular bonds. It is noteworthy that this shielding effect results, in the case of tertiary amyloxide groups, in the formation of a monomeric zirconium derivative which boils at a temperature three degrees below the boiling point of the corresponding monomeric titanium compound. This surprising phenomenon of the substance of higher molecular weight being the more volatile was further illustrated by the discovery that hafnium tertiary amyloxide was more volatile than the zirconium compound (table III).

TABLE III

Compound	*B.p./0·1 mm*	*Mol. weight*
$Ti[OC(CH_3)_2C_2H_5]_4$ ..	98	396·5
$Zr[OC(CH_3)_2C_2H_5]_4$ ..	95	439·8
$Hf[OC(CH_3)_2C_2H_5]_4$ ..	92	527·2

This interesting behaviour is of considerable theoretical interest, and further investigation [19] has shown that the phenomenon is not confined to metal alkoxides.

The stereochemical theory advanced to account for the physico-chemical properties of the alkoxides of titanium, zirconium, and hafnium would lead

TABLE II

Alcohol	*Titanium alkoxide*			*Zirconium alkoxide*		
	B.p./mm	*M*	*X*	*B.p./mm*	*M*	*X*
$CH_3.CH_2.CH_2.CH_2.OH$ ..	175/0·8	565	1·4	256/0·01	1428	3·2
$(CH_3)_2CH.CH_2.OH$	148/0·1	490	1·2	247/0·1	1462	3·3
$(C_2H_5)(CH_3).CH.CH_2.OH$ ..	154/0·5	422	1·1	238/0·1	1615	3·7
$(CH_3)_3C.CH_2.OH$	105/0·05	500	1·3	188/0·2	1051	2·4
$(C_2H_5)_2CH.OH$	112/0·05	399	1·0	178/0·05	868	2·0
$(C_3H_7)(CH_3)CH.OH$ ..	135/1·0	410	1·0	175/0·05	860	2·0
$(CH_3)_2CH.CH(CH_3).OH$ ..	131/0·5	415	1·0	156/0·01	870	2·0
$(CH_3)_2(C_2H_5).C.OH$ ..	98/0·1	405	1·0	95/0·1	455	1·0

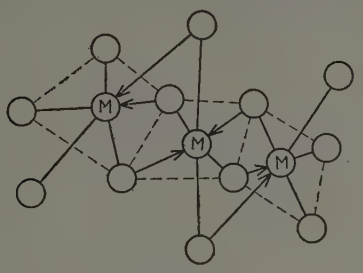


FIGURE 2

one to predict that the shielding requirements of a metal are determined by its atomic radius. Thus it appears from table II that the shielding requirement of titanium is smaller than that of zirconium, and accordingly it was expected that the requirement of thorium would exceed that of zirconium. This was indeed found to be so, because thorium gave a complex tertiary butoxide and a complex tertiary amyloxide, in contrast to the analogous monomeric compounds of titanium, zirconium, and hafnium. With thorium it was necessary to prepare the derivatives of triethyl carbinol $(C_2H_5)_3C.OH$ or methylethyl*iso*propyl carbinol in order to obtain monomeric compounds. The dependence of shielding requirement on radius of the central atom is clearly revealed in table IV, where the boiling points (° C at 0·1 mm pressure) and molecular complexities (in brackets) of a range of compounds are given.

TABLE IV

Central Atom ..	*Ti*	*Zr*	*Th*
Atomic radius (Å) ..	1·36	1·48	1·65
Alkoxide:			
$CH_3.CH_2.O$ ..	102(2·4)	180(3·4)	Non volatile
$(CH_3)_2.CH.O$ ..	49(1·4)	160(3·0)	210(3·8)
$(CH_3)_3.C.O$ ..	52(1·0)	50(1·0)	160(3·4)
$(CH_3)_2(C_2H_5)C.O$	98(1·0)	95(1·0)	200(2·8)
$(CH_3)(C_2H_5)_2C.O$	128(1·0)	128(1·0)	148(1·8)
$(C_2H_5)_3C.O$ ..	166(1·0)	166(1·0)	154(1·0)

These monomeric alkoxides of thorium are more volatile than any other derivatives of this element. It seems possible that these volatile derivatives of the metals may have important industrial applications where high purity is required. Preliminary experiments show that distilled zirconium alkoxides are spectroscopically pure with respect to many common metallic impurities. Moreover, it is possible to separate hafnium from zirconium by high-efficiency fractional distillation. A new method for the separation of niobium and tantalum by means of their alkoxides is also foreshadowed. The ability to prepare the alkoxides in a high state of purity and the ease with which they can be converted to the pure oxide is of great value, particularly when the metal can be obtained by reduction of the oxide.

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Petrology as an aid to prehistoric and medieval archaeology

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Some two years ago we surveyed in an editorial (ENDEAVOUR, 12, 59, 1953) the rapidly increasing contributions being made to archaeology by the other sciences. In Britain, this hitherto informal co-operation was greatly strengthened last year by the formation of a special committee of the Council of British Archaeology for the purpose of co-ordinating the application of scientific research. In this article Dr F. S. Wallis describes some valuable results obtained by the application of petrology to archaeology—for example in locating ancient axe-factories and the trade routes by which their products were distributed.

In any discussion of the contributions of petrology to archaeology, the careful and pioneer investigations by the late H. H. Thomas [1], in 1923, of the source of the megalithic stones at Stonehenge will always remain a classic. After other workers had contributed what were little more than inspired guesses he showed the great value of precise investigation. His unequivocal identification, by petrological methods, of the source of the blue stones of Stonehenge as the Presely Mountains[1] in Pembrokeshire gave archaeologists a firm basis for speculation about the way in which these massive stones were transported to their present position and other problems relating to the construction and function of this famous monument.

A decade later, the group of museums and art galleries in the south-west of England set up a committee to investigate the original provenance of rocks used by Neolithic and Early Bronze Age man in making artifacts, particularly stone axes. Three reports have been issued [2], and the work, which we shall discuss in some detail later, has proved to be of such value that it is now on a national instead of a local basis.

In 1938 F. J. North [3] summarized the archaeological advantages to be gained from an exact knowledge of the petrology of materials and gave many striking examples of problems which had been elucidated by a knowledge of the rocks concerned. More recently still, W. J. Arkell [4] has written a valuable book on the origin of the materials used in the ancient buildings of Oxford, and his work has already proved an incentive to similar studies elsewhere. In particular, E. M. Jope [5] has made some brilliant architectural and economic researches and has clearly shown the fundamental value of exact petrological identification.

The inter-relationships of archaeology and geology are thus many and varied, and they are, in a broad sense, complementary to each other in the elucidation of the stirring story of the Earth and its inhabitants. It is becoming increasingly realized that archaeologists and economic historians must collaborate with petrologists in the solution of many problems.

PETROLOGY OF PREHISTORIC SITES

In former days archaeologists ignored the underlying solid geology of the sites they investigated, though they gave some attention to the nature of gravels and other superficial deposits. Gradually this policy has changed, and no excavation is now considered to be complete unless the geology of the area has been studied and the petrological character of all finds of stone and pottery has been determined. An interesting example may be quoted from Pany-y-Saer, Anglesey [6], where C. W. Phillips found some rough, but rather remarkable, pottery in the remains of some buildings occupied between the 2nd and 4th centuries A.D. The mineral fragments in the crushed pottery were examined, and the nature of these proved conclusively that they had originated in the local Boulder Clay. It was also shown that these early people used the Upper Boulder Clay, which in Anglesey occurs in two varieties—blue or brownish-grey, and red. Thus the petrological evidence proves that the potters blended two local clays in making their final mixture—a fact of considerable social and economic importance.

In America, further work of this type has been undertaken [7] and the possibility of examining

[1] For the location of this and other places mentioned in the article see figure 1.

FIGURE 1

thin sections of sherds of pottery, instead of isolating the heavy minerals, is being investigated. The study of thin sections has been used to obtain valuable information regarding the amount of tempering material added to the original clay base and the temperatures reached in firing.

On some archaeological sites the nature of the underlying rock has provided evidence about the water supply available to the prehistoric inhabitants. Geological evidence has also permitted interesting deductions about the original siting of dwelling places and fortifications.

EXAMINATION OF STONE AXES

Archaeologists were originally interested only in the form and stratigraphical position of artifacts such as axes. The importance of a proper petrological examination of the numerous stone axes in public and private collections has, however, been stressed for many years past. It was urged that a knowledge of the distribution pattern of these axes would lead to a far wider and more exact understanding of the early trade routes and other factors of economic and social importance in Neolithic and Early Bronze Age times. Fortunately there is no lack of material; many such objects have been preserved in Victorian drawing-room cabinets, in the belief that they were charms or thunderbolts. A major hindrance to research, however, lay in the natural reluctance of owners to permit any damage to the specimens. Macroscopic examination, however, was rarely fruitful, for the grinding, polishing, and patination which had taken place during and after manufacture had generally obliterated any diagnostic features. It was therefore essential for the petrologist to be able to make his diagnosis by the study of a thin section under the microscope. A technique was evolved for cutting a small, plate-like piece from the stone and then grinding it to the required thickness. The

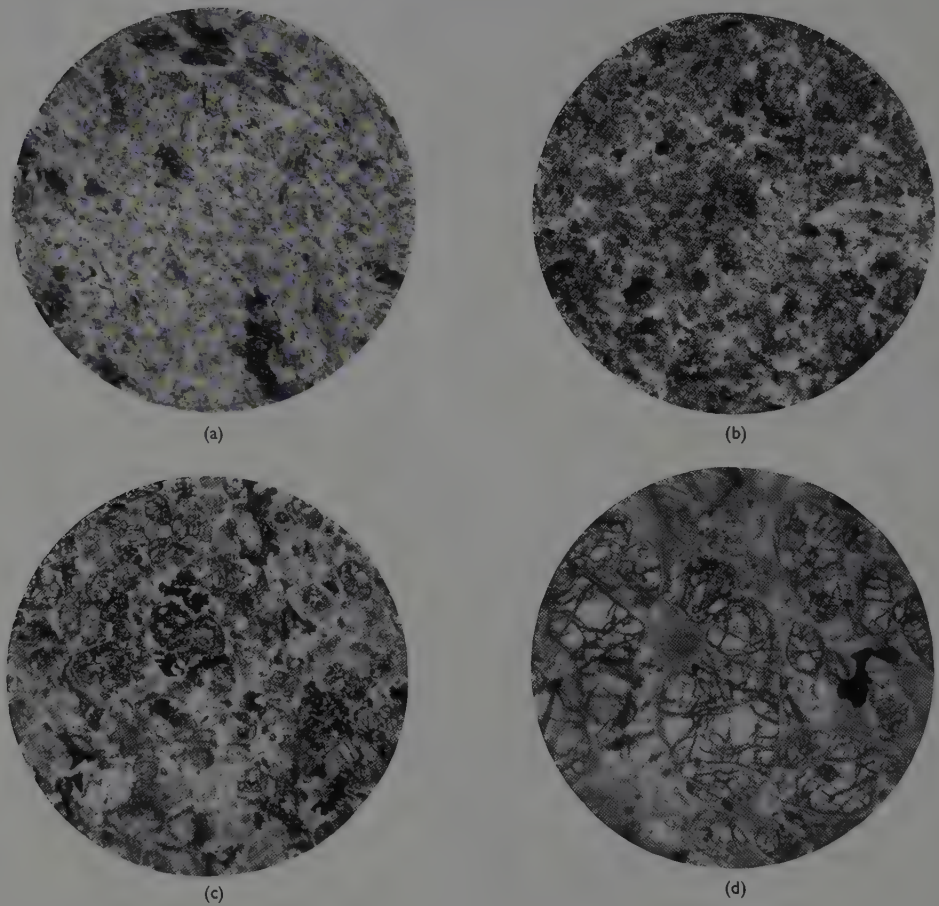

FIGURE 2 – *Photomicrographs of sections, in ordinary light, of axes, with corresponding rocks for comparison.* (a) *Group VII axe, from Avebury, Wiltshire.* (b) *Rock from Graig Lwyd.* (c) *Group XII axe, from Rollestone, Wiltshire.* (d) *Rock from Cwm-mawr, Montgomeryshire.* (*All* × 18) *(Photographs by J. Rhodes, E. Seavill, and F. W. Shotton)*

small cavity left in the object was then filled with plaster and coloured. Thus from the collector's standpoint the stone was unaltered in appearance, and yet it had yielded the maximum petrological, and hence archaeological, information.

Many hundreds of axes have been examined in this way. Realizing the difficulty of immediately assigning an original provenance to the rocks, it was decided to allocate them to well-defined but arbitrary groups. Some seventeen such groups have already been recognized, and it is probable that the number will increase with the increase in the number of workers in this specialized but interesting field.

Groups I–IV are greenstones of varying types. Group I is a well-defined rock which weathers to a characteristic rough surface, and may be technically termed a uralitized gabbro. In this rock augite, sometimes slightly ophitic, occurs in colourless to pale-brown plates, which show a variable amount of alteration to green, fibrous hornblende. It is the presence of this secondary amphibole which accounts for the extremely tough nature of these axes. The felspars, probably albite, are much altered and are more or less riddled with needles of actinolite. Ilmenite is the chief accessory mineral. All geologists know the extreme variability of greenstones even within a

small outcrop; the great uniformity which characterizes this group of axes shows that the prehistoric men who made them must have collected their material from a very small area. Unfortunately, the exact provenance of this rock still eludes the investigators, though a rock of very similar composition to that of the axes of Group I has been found at Gear Rock, in Mount's Bay, Cornwall. In a review of the evidence for the submergence of southern Britain, Glyn Daniel concludes that in Megalithic times the coastline was at what is now the 5-fathom line. It thus appears likely that the original source of Group I axes—and this important group comprises more than one-tenth of all the axes examined—was in Mount's Bay. The products of this factory are typologically very varied; originality in design and a breakaway from tradition were the keynotes of this relatively large industrial undertaking of Neolithic times.

Group II greenstone axes are relatively few in number, but their point of origin can be confidently assigned to a site on the coast between Lay Point and St. Ives, Cornwall. Group III is another small group, deriving from a neighbouring site at Trenow Quarry, Marazion, also in Cornwall. Here some neighbouring crags would probably give evidence of a factory if a systematic investigation were made.

On petrological evidence Group IV has been located at Balstone Down, Callington, Cornwall; the rock of which these axes are made is easily recognized in thin section as a sheared tremolite with chlorite and ilmenite. This was a relatively small factory, making chiefly normal axes, but with a tendency to specialize in large axes with thin and nearly pointed butts.

Group V is of little importance, but Group VI comprises axes which were at one time thought to be made only at Stake Pass in the Lake District, but are now known to be characteristic of the larger Great Langdale area. The rock is an epidotized tuff of intermediate or basic composition, belonging to the Ordovician (Borrowdale Volcanic series). B. Bunch and Miss C. Fell [8] have graphically described this important axe factory, the focus of which was on the scree slopes of Pike of Stickle. As at Graig Lwyd and at Tievebulliagh Hill, near Cushendall, County Antrim, which will be discussed later, it was the loose screes which attracted the early craftsman and gave him his raw material. As no polished axes have been found on the site, it is clear that only rough flaking was carried out at Great Langdale and that the finishing processes were done at one of the more permanent all-the-year-round settlements, such as Ehenside Tarn. Axes from the Pike of Stickle were traded over a wide area, for they have been found in Cumberland, the Midlands, south-west Scotland, the Isle of Man, and County Antrim, as well as the extreme south of England. As for other factories, coastal and riverine routes were favoured for distribution.

A well-known factory site on the crags of Graig Lwyd, overlooking the sea on the northern slopes of Penmaenmawr, North Wales, yielded the axes of Group VII. The rock is an augite granophyre (figure 2) and is extremely tough; it flakes well and gives good bulbs of percussion and fairly smooth conchoidal fractures. As at Great Langdale, the craftsmen concentrated on the manufacture of plain axes. The distribution pattern of these axes indicates that they were traded to Wiltshire through the Midland gap. An interesting small sub-group, VIIa, in which the rock is similar to, but not identical with, the Graig Lwyd granophyre, has recently been shown by P. R. Giot to be the exact equivalent of his Breton A group. The British examples came from Bournemouth, Southampton, Tewkesbury, and the Channel Islands, pointing to a cross-channel trade from France to ports on the south coast of England. Unfortunately, the source of the French group cannot yet be located more exactly than as being in Brittany, but it is certainly from the Armorican massif.

Groups VIII and XI may be described petrologically as silicified rhyolitic tuffs, though their exact differentiation is still a matter for discussion. Present indications rather point to the source as being outcrops of Llanvirian volcanic rocks (Ordovician) on Ramsey Island and on the mainland at a quarry near Mountjoy, Pembrokeshire, for Group VIII, according to a suggestion made by Mrs J. Morey [9]; and to the Great Langdale area for Group XI. The former suggestion has recently received support from the finding of an axe of Group VIII at Caldy Island, though this has been questioned, on typological grounds, by P. R. Ritchie [10].

Rock from the screes of Tievebulliagh Hill is the source of Group IX axes. It is a hard, mottled, bluish-black rock with darker spots, has a flinty aspect, and breaks with a splintery fracture. It is called porcellanite. It is composed of dark spinel and a light fine-grained aggregate of sillimanite and mullite. E. M. Jope [11] has

described these factories in some detail, and has shown that in addition to a well-known one at Tievebulliagh there was another on Rathlin Island. As in the case of both Great Langdale and Graig Lwyd, no polishing appears to have been carried out at either of these two Irish factories. As no rough forms have been found outside County Antrim, it would appear likely that only polished axes were traded to Britain.

Group XII is a particularly interesting one, because it can be definitely related to a small area at Cwm-mawr, just south of Corndon Hill, Montgomeryshire [12]. This factory specialized in large perforated axe hammers—a type that is sometimes found in Group I. The rock is a picrite (figure 2) of very definite composition. The occurrence of this rock is limited in Britain, and its identity with the small outcrop at Cwm-mawr seems assured. The distribution pattern of axe-hammers found shows a preponderance in the West Midlands, with a concentration around Cwm-mawr, but it also occurs as far afield as Avebury, in Wiltshire, and Otterton, in Devon. This confident identification of rock material as coming from such a relatively small area awaits final confirmation by excavation of the factory site. From petrological and topographical considerations F. W. Shotton believes that a locality on the south-eastern side of the hill is the most likely site; this is a small, pleasant place with flat ground immediately below which would have been appropriate for the workshop.

Group XIII contains axes made from rock outcropping at Carn Meini and Cerrig Marchogian in the Presely Mountains of Pembrokeshire. This handsome and distinctive rock, with white or pinkish felspathic spots, was used for constructional purposes at Stonehenge, and is also found, as axes and axe-hammers, in such widely separated areas as County Antrim, Wales, Windmill Hill in Wiltshire, and Sidmouth in Devon. Unfortunately the exact site of the factory still eludes the archaeologists.

In recent months four other groups have been established. The essential petrological feature of Group XIV is a deep-brown pleochroic hornblende occurring in large patches, which may have good outlines, though it is frequently ophitic. There is also some augite, turbid plagioclase felspar, apatite, sphene, and pyrites. The group has been matched with a camptonite from Griff, near Nuneaton. Shotton, in addition to the determination of Group XIV, has identified a series of axes classified as Group XV; these are made of a particular type of greywacke. The rock is a chloritic, sericitic siltstone.

Groups XVI and XVII comprise two distinctive greenstones. The former is characterized by the presence of considerable quantities of hornblende occurring as patches of felted green needles, often with a parallel arrangement. It has been matched with greenstones from the Camborne area of Cornwall. Group XVII axes are made from rock in which parallel folia of clear, crystallized felspar occur in the form of a mosaic; between these are layers of brownish, fibrous hornblende, often containing knots around which the felspar folia are bent to produce augen structures. The magnetite occurs as uniform grains in the larger hornblende masses, giving the rock a characteristic appearance. Similar rocks occur near Kenidjack Castle and other places in West Cornwall.

As is to be expected from consideration of the geological structure of Britain, it was the northern and western districts which supplied the tough igneous rocks used for the manufacture of these early implements; in the eastern and southern areas flint was mainly used, and the mines there bear witness to the source. It is interesting to note that in a recent examination of some stone axes from East Anglia examples of only the more successful groups—namely Graig Lwyd, Great Langdale, and Group I—were encountered. These were the more popular types, and in consequence were traded to distant parts of the country.

From this intensive research, now being carried on in several parts of Britain, there is slowly emerging a picture of the trade routes involved in the economic dispersal of early tools and weapons. The distribution pattern clearly indicates the great importance of coastal and riverine routes, though the products of the Graig Lwyd factory were mainly traded overland through the Midland gap.

The importance of Avebury in Neolithic times is clearly revealed by this new line of research, for it is shown to have been the centre of the axe trade. Products of all the important factories, and even of most of the lesser ones, are represented there, and Niedermendig lava from the Rhine has also been recorded. This is an impressive fact, and it has been suggested that a possible explanation is to be found in some form of spiritual rather than material exchange, for it is difficult to believe that there can have been any considerable bartering at Avebury with products of the local chalk and flint.

PETROLOGY AND MEDIEVAL ARCHAEOLOGY

The growing value of petrology to the prehistoric archaeologist is thus evident, but it is encouraging to see that scholars interested in later periods also recognize that an exact determination of the nature of stone used can assist the elucidation of the complete story. Thus the patient determination of the several types of stone used in Oxford, by W. J. Arkell [4], has made an important contribution to the study of the regional building characteristics, now rapidly disappearing. The source of the many types of stone used in Oxford has been traced back to the individual quarries from the Middle Ages to the present day. It is academically a valuable survey and has also a practical value, for it has long been established that the most aesthetically satisfactory repairs and restorations of buildings are those in which the original type of stone is used rather than those now fashionable, however sound and durable these may be.

Similar studies are being made elsewhere. For example, granite obtained from the five outcrops in the south-west of England was used in both Cornish and Devon churches in rebuilding which took place in the fifteenth and sixteenth centuries, and the distribution pattern of the easily recognized Purbeck marble is slowly becoming clear.

Recently, E. M. Jope and G. C. Dunning [13] have carried out a survey of the use of blue slate for roofing in medieval England. This is based on petrological evidence linked with documentary records. The result, though not final, is illuminating, and indicates that, with the exception of a Belgian slate found in the deserted medieval village of Hangleton, the roofing slates found in many medieval sites in southern England are from the Devonian formation. Not only has it been possible to establish the sites of the old quarries, but the transport routes on the southern coast, for example from Fowey to Southampton, have been worked out. The routes of the blue slates from other localities in the western parts of Britain, including Pembrokeshire and North Wales, have also been traced; these too indicate a vigorous coastal trade. The nature of the export trade to Ireland, France, and the Netherlands has been established in broad outline, emphasizing the fruitful results that may be obtained by collaboration between the economic historian and the petrologist.

In April 1954 an informal conference of archaeologists and scientists was held in London to discuss how they could best help each other. Later in the year this led to the formation of a Scientific Research Committee of the Council of British Archaeology, set up for the purposes of co-ordinating and advising on the applications of scientific research to archaeology. Collaboration between the petrologist and the archaeologist, with particular reference to the study of stone axes, is one of the major considerations of this committee.

In preparing this summary I have drawn freely on the work of colleagues, and I should like to take this opportunity of acknowledging the consistent co-operation of Mr E. D. Evens, Mr E. M. Jope, Mrs J. Morey, Professor F. W. Shotton, and Dr J. F. S. Stone.

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The permanent magnetization of rocks

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There is strong evidence, here discussed, that the direction of magnetization of rocks perpetuates that of the Earth's field at the time of their formation. On this assumption it is possible, by examining rocks of known age, to discover the variations which have taken place in the direction of the Earth's field. Of outstanding interest is the considerable body of evidence that the geomagnetic field has in the past undergone repeated reversals.

The fossilized remains of once living organisms indicate the conditions of life obtaining at the time of their formation. Recently it has been found that there exists another, equally widespread, fossil—but of a physical rather than biological nature. It has been shown that many sedimentary strata, or series of lava flows or other igneous rocks, are magnetized in various directions, but that in general the directions are uniform within a given geological epoch. These magnetic fossils can be of considerable value.

The only common minerals capable of becoming permanently magnetized are the iron oxides; chief among these are the titanomagnetites and haematite. The former are a series of solid solutions of magnetite (Fe_3O_4) and ulvospinel (Fe_2TiO_4). In this series the Curie point (above which ferromagnetism disappears) is lowered, and the saturation magnetization decreases, as the proportion of titanium increases; the Curie point for pure magnetite is 575° C, but it is 220° C if the molecular proportion of ulvospinel is 50 per cent. Haematite (α-Fe_2O_3) has a Curie point of 675° C, and a saturation magnetization of about 0·5 e.m.u./g, compared with 92 e.m.u./g for pure magnetite. Haematite forms solid solutions with ilmenite ($FeTiO_3$), and these are ferromagnetic. Maghemite (γ-Fe_2O_3) has a cubic lattice similar to that of magnetite and is strongly ferromagnetic, but as it is unstable at temperatures above approximately 300° C it is likely to have resulted in the main from the oxidation of magnetite during the weathering of rocks.

These iron oxides are not easy to study by the usual petrological techniques, as they are opaque even in thin sections: consequently their wide range of composition was not appreciated until comparatively recently. A further complication is that structures occur in which laminae of one component have separated out from solid solutions; these may form intergrowths, of two different compositions, ranging in thickness from a few microns upwards. With the metallurgical microscope these intergrowths may be seen on the polished surface of the ore grains, especially if the latter are etched. However, the structure and texture of iron oxide minerals, which form only a few per cent of the material studied, are of considerable importance to the magneticians interested in the remanent magnetization of the rocks, so that study of detail may be essential.

There seem to be two ways in which such minerals can become magnetized. If an unmagnetized assemblage of grains, above the Curie point, is cooled down in a magnetic field, even one as weak as that of the earth (0·5 gauss), the grains can acquire a magnetization corresponding to a considerable fraction of their saturation magnetization. This thermo-remanent magnetization (T.R.M.) arises because just below the Curie point the coercive force is only a fraction of a gauss. Provided the grain size is neither too large nor too small, however, the coercive force at ordinary temperatures is large, 50 gauss for pure magnetite and several thousand gauss for haematite. T.R.M., when once acquired by a rock, is stable, because rocks are not generally subjected to strong magnetic fields nor to high temperatures. The second way in which such grains may become magnetized depends upon the fact that at ordinary temperatures some iron oxides may acquire appreciable isothermal remanent magnetization (I.R.M.) by prolonged influence of the geomagnetic field. E. Thellier and F. Rimbert [1] have shown that I.R.M. is more easily destroyed by alternating-current demagnetization than is T.R.M.

If a rock is magnetized in the same direction as the present geomagnetic field, it may be either because the field was in the same direction at the time of the rock's formation, or because the rock has acquired appreciable I.R.M. over the last few

hundred thousand years. In the latter case the rock is regarded as magnetically unstable, and it is distinct from those which have successfully 'fossilized' the geomagnetic field existing at the time of their formation. There is at present no infallible test for instability, but it is prudent to exclude rocks with low coercive force or low Curie points and those which are lithologically similar to rocks known to be unstable.

Proof that T.R.M. is responsible for the natural magnetization found in a rock is in principle not difficult. The natural magnetization should decay, as the rock is heated, in exactly the same way as does the T.R.M. given to another sample of the same rock by cooling it from above the Curie point in the geomagnetic field. This is an important way of investigating which of the iron oxides are present, because the decay of magnetization is sufficiently rapid just below the Curie point to allow this temperature to be determined quite accurately. It is unfortunate, however, that not only may a naturally occurring rock have lost some of its magnetism through ageing, but that the geomagnetic field may be of a very different strength today from that obtaining at the time of formation of the rock. Consequently the heating experiments may give only qualitative evidence to support the presumption of the thermo-remanent origin of the magnetization. The problem may indeed be more considerable than this; on heating, changes may occur in the opaque minerals which alter their composition and Curie points. In this case we shall get no evidence whatever of the nature of the initial process of magnetization.

It seems, however, that intense and stable T.R.M. probably accounts for the natural magnetization of all the rocks which are of interest in the present context. The rocks of the Earth's crust have been formed either directly by the cooling of molten magma to form igneous rocks, or by the deposition, and subsequent consolidation and cementation, of eroded fragments of other rocks, forming sediments which are usually well stratified. Both igneous and sedimentary rocks may—by deep burial under pressure and at high temperature, or by chemical action—be appreciably changed. The magnetization of such metamorphic rocks has not so far been much investigated, as their history is clearly complex. Going back in geological time, one would naturally expect to find more strata in which the original magnetism has been lost and which have become wholly or partially remagnetized along new directions.

MAGNETIZATION OF IGNEOUS ROCKS

In the present context the most interesting examples of igneous rocks are perhaps the lavas, which often emerge from fissures in the ground. If basaltic, and therefore of low viscosity, they can flow over large distances, perhaps tens of miles, and form layers of remarkably uniform thickness, often some dozens of feet thick. In some places successive eruptions of lava dominate the landscape, as in the Columbia River region in the north-west of the United States, and in Iceland. In such a region one has a clearly defined series of rocks which not only must have acquired T.R.M. but form a chronological sequence, the intervals between eruptions ranging from a few years to a few tens of thousands of years.

In a series of measurements of the Tertiary basalts in Iceland, J. Hospers [2], found that some groups of lavas are magnetized in the present direction of the geomagnetic field and other groups in exactly the opposite direction. This normal and reverse magnetization has now been found to be characteristic of the Columbia River basalts (C. D. Campbell and S. K. Runcorn [3]) and of those of the Central Massif of France (A. Roche [4]), all of which are of Tertiary age.

Isolated examples of reversed magnetization had previously been noted. In particular, it was known from magnetic surveys that many dykes—walls of solidified magma which were once fissures through which molten lava had issued—consisted of reversely magnetized material. Figure 2 shows an example of a dyke cutting through horizontal lava flows in the Columbia River basalts of the north-west United States. H. Gelletich [5] demonstrated that the great Pilansberg system of dykes in South Africa was reversely magnetized, and J. M. Bruckshaw and E. I. Robertson [6] showed that the magnetization of the tholeiite dykes of northern England also was reversed.

The interpretation of these results has led to much discussion. Those who have examined the rocks in question see no reason to doubt that, in general, reversed magnetization arises from a reversed geomagnetic field existing at the time of cooling of the lava. The fact that reversals occur a number of times in the series of lavas mentioned above is taken to indicate that reversals of the geomagnetic field may take place at irregular intervals, of the order of a few hundred thousand to a few million years.

P. M. S. Blackett [7] has recently called this explanation of reversed magnetization in rocks the 'obvious' one. On the other hand, as recently as

1951 L. Néel [8] could preface a paper, which attempted to show how reversed magnetism could arise from anomalous properties of the iron oxide minerals, with the statement that the theory of the geomagnetic field precluded reversals. It is only within the last few years that the idea of reversals of the main geomagnetic field has come to be seriously entertained by physicists as a likely explanation of reversed magnetization. The growth of our concepts of the physical processes which go on in the Earth's interior has had much to do with this growth of our ideas about the geomagnetic field. These concepts are twofold. The Earth possesses a liquid core some 3400 km in radius; the comparatively rapid changes in the geomagnetic field, indicated by the variation of the magnetic declination in Europe during the last five hundred years, point to fluid motions in this core, probably the result of thermal instability. The gyroscopic effect of the rotation of the Earth is a dominant factor in the symmetry of these motions, and results in the coincidence of the magnetic axis of the Earth with its axis of rotation when the effects of the secular variation are averaged over a few thousand years. Without going deeply into the theory of the main geomagnetic field, it is evident that on this basis we can attach no fundamental significance to its polarity. Indeed, all attempts to produce a satisfactory explanation of the field make it evident that the external dipole field is secondary to, and much smaller than, the so-called toroidal field, the lines of force of which remain within the electrically conducting interior of the Earth.

Nevertheless, a number of attempts have been made to explain the anomalous magnetization in terms of unusual properties of the iron oxide minerals rather than by reversal of the geomagnetic field. Néel [8] has shown that four such possibilities exist. Two processes apply to igneous rocks, and relate to the method by which magnetization is acquired on cooling. If the iron oxide minerals consist of two separate ferromagnetic phases, as in the intergrowths previously discussed, and if a phase A has a higher Curie point than phase B, it is possible that when its Curie point is reached B will be cooling in a field reversed by the 'return field' of the already magnetized material A. If A has a smaller temperature coefficient of magnetization than B, it is possible for the reversed magnetization of B to be predominant at ordinary temperatures. T. Nagata [9] has apparently found two volcanic rocks which behave in this way, but careful examination of the basalts referred to above has revealed no such properties in them. The second process suggested by Néel depends on the imperfect antiferromagnetism of the iron oxides. Owing to quantum mechanical exchange forces, the magnetic moments of the atoms in a domain in iron are aligned in parallel and in ferrous oxide in antiparallel. Thus the latter material does not have ferromagnetic properties and cannot be permanently magnetized. The ferromagnetism of such a material as magnetite is due to an imperfect antiparallel arrangement of elementary moments, those of one sign outweighing those of the other. Néel shows that in some cases this situation may be reversed by change of temperature, and a lithium ferrite has been synthesized that has this property, but no naturally occurring rock is known to possess it.

The other processes which Néel suggested apply both to lavas and sediments, but have not been experimentally demonstrated. These suppose that the two processes described above may occur at ordinary temperatures as a consequence of chemical or structural changes in the iron oxide minerals, occurring perhaps only over millions of years.

It is possible that such phenomena must be taken into account in the interpretation of reversed magnetization. Néel's processes seem, however, to depend on coincidences too improbable to allow their very widespread occurrence in nature. Moreover, no differences have been found in average magnetic properties between reversed and normal specimens. Nor do the two processes described above which have actually been observed account for reversed magnetizations in sediments which have been investigated in the last few years.

MAGNETIZATION OF SEDIMENTS

To extend the palaeomagnetic record, particularly for pre-Tertiary times, it is necessary to examine the remanent magnetization of sediments as well as of lava flows. Work of this kind has been going on for some twenty years, but it is only recently that interesting and significant results have been obtained. In general, the intensity of magnetization of lava specimens lies between 10^{-2} and 10^{-4} gauss, while that of sediments is less than 10^{-4} gauss. The smaller proportion of iron oxides in sediments does not alone account for this difference, which must therefore be due to the iron oxide grains having a smaller intensity of magnetization than those in lavas, or to a much greater scatter of the directions of magnetization. The iron oxide grains must have originated in an

FIGURE 1 (*above*) – *Red Sandstones and shales of the Supai (Permian) formation of the Grand Canyon, Arizona.* (a) *Kaibab limestone;* (b) *Coconino sandstone;* (c) *Supai formation (shales and sandstones);* (d) *Redwall limestone.*

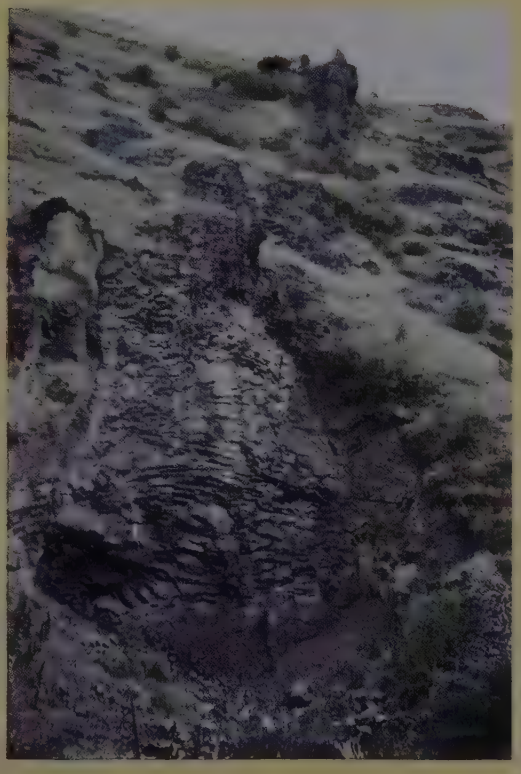

FIGURE 2 (*right*) – *Dyke cutting through horizontal lava flows in the Columbia River basalts, near Redmond, Oregon.*

FIGURE 3 – *Astatic magnetometer, showing Helmholtz coils, for removing the horizontal and vertical components of the geomagnetic field, the magnetometer, and specimen holder.*

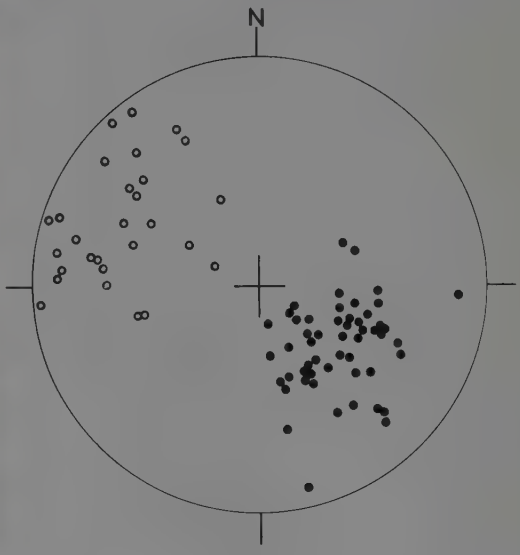

FIGURE 4 – *Directions of magnetization of Torridonian sandstones. Open circles = upward directions. Closed circles = downward directions.*

igneous rock, in which they presumably had T.R.M. We might expect the process of erosion, transport, and deposition to which the grains would be subjected, and possible chemical changes also, to reduce this magnetization very considerably. At any rate, it is significant that when a sediment is heated beyond the Curie point and then cooled in the geomagnetic field, it can receive a magnetization a thousand times its natural one. We may tentatively believe that during the deposition of the sediment in reasonably still water the magnetized iron oxide grains rotated under the influence of the ambient geomagnetic field, giving the sediment a weak magnetization in the direction of the field.

Figure 3 shows the astatic magnetometer used at Cambridge for the measurement of the magnetization of sediments: it is a development of the astatic magnetometer made by P. M. S. Blackett [10]. The specimen, in the form of a short cylinder, is raised underneath the lower magnet of the astatic pair and the deflection is noted. The procedure is repeated with the cylinder rotated through successive small angular increments. In this way one can measure the component of the magnetization in different directions in the plane of the disc. If the disc is cut out of the rock specimen in such a way that its axis corresponds to what was the vertical when the sediment was laid down, these measurements give the palaeomagnetic declination. A more complicated procedure enables the inclination to be measured also.

The first sedimentary series to be examined in Britain were rocks from the Torridonian sandstones of north-west Scotland; these are of late pre-Cambrian age. Sixty-three localities, spread over seventy miles of the coast, were sampled by E. Irving, of the Department of Geodesy and Geophysics, Cambridge University, to provide material for an examination of a reasonable fraction of the succession, which is 3000 m thick. The reversals of magnetization so characteristic of the Tertiary lava flows are found also in this series of rocks. Reversals occur every 30–600 m in the succession. What this means in terms of geological time is, of course, impossible to determine, but a mean rate of deposition of 1 cm per year is probably of the right order of magnitude. Ten localities have been found in which directions of magnetization occur which are widely different from those in fifty-three other localities. These variant directions of magnetization are thought to represent periods during which the geomagnetic field was in the process of reversal. During such a period of change one would not expect the field at the Earth's surface to be a simple dipole: one would expect a much more complicated field. Consequently the presence of these oblique zones

gives support for the reality of reversals of the geomagnetic field. The magnetization of these rocks differs from those of the Tertiary age in that the directions are at a large angle to those of the present field; this is shown in the accompanying stereogram (figure 4).

FIELD TESTS OF MAGNETIC STABILITY

The evidence that the direction of magnetization of sediments is indicative of the direction of the geomagnetic field at the time of deposition is not very direct. It is possible, by making use of some ideas put forward by J. W. Graham [11], to show that the magnetization has not changed since early in the history of the rocks. Two tests of the permanence of magnetization acquired at the formation of a rock are afforded by nature. First, in folding and other earth movements, strata may be so moved that contemporaneous layers lie for much of geological history with their bedding planes at very different angles in different outcrops. If there is any tendency for such rocks to be magnetically unstable, even over periods far too long to be observable in the laboratory, they will become magnetized partially or wholly in the direction of a magnetic field imposed later. In that case the directions of remanent magnetization found in the rocks today will be more nearly parallel than one would expect if their magnetization had been assumed on formation. The second field test of stability consists in examining the remanent magnetization of conglomerate pebbles, derived from the rock strata through erosion. In the formation of the conglomerate bed the pebbles would obviously be deposited without reference to their magnetization, i.e. the directions of magnetization would be random. Unless the material from which the pebbles were formed is magnetically unstable, this randomness will persist.

The above tests do not exclude the possibility that the original magnetization has assumed some preferred direction within the rocks. It might be, for example, that iron oxide particles of a particular shape are preferentially orientated by hydrodynamic forces and by gravity during deposition. It is possible that such particles retain their magnetization better than those of irregular shape, in which case the direction of magnetization in the rock would gradually change. If such effects do occur they are probably small, but corrections may have to be made for them. Perhaps the most cogent argument against them is the uniformity of magnetization found over great distances in such a series as the Torridonian. It is even more convincing when igneous rocks and sediments of the same age can be compared.

SURVEY OF THE MAGNETIZATION OF THE GEOLOGICAL COLUMN

Much progress has been made in finding the directions of magnetization characteristic of a considerable number of geological epochs. K. M. Creer, of the Department of Geodesy and Geophysics, Cambridge University, has selected Red Sandstones (of the type that Irving found to be stable in the Torridonian) from a number of geological epochs and found them to be consistently magnetized within each epoch. These directions are given in table I, and are taken from a brief paper by Creer, Irving, and Runcorn [12]. Also, an extensive survey of the New Red Sandstone of Triassic age has been made by J. A. Clegg, M. Almond, and P. H. S. Stubbs [13], who show that these rocks are consistently magnetized along a NW–SE axis. Approximately equal numbers of reversed and normal magnetizations were found.

Well-bedded fine-grained Red Sandstones of the type which have been extensively used in this work occur in many geological formations and are well exposed at many levels in the Grand Canyon, Arizona (figure 1) where work is being undertaken at present.

POLAR WANDERING

It is clear that some unifying hypothesis is required to give significance to these varying palaeomagnetic declinations and inclinations. If one considers the situation that obtained in Tertiary times, one sees that in spite of frequent reversals the direction of the magnetization of the lavas remains close to that of an axial dipole. J. Hospers [14] has considered this point carefully. Modern palaeontology is quite definite that no appreciable change in the position of the pole could have occurred in Tertiary times. Thus in order to explain the pre-Tertiary results it seems reasonable to suppose that the axis of rotation and the magnetic axis have always coincided, an assumption which has already been justified theoretically. It is therefore necessary to assume that there has been a movement of the land relative to the poles. This possibility has long been seriously considered by geologists in order to explain certain climatological and palaeontological difficulties.

It should be made clear that there are two possibilities to distinguish between. Continental drift envisages the separation of the continents, while

TABLE I. *Direction of magnetization of Red Sandstones*

Geological period	*Approximate age (million years)*	*Average directions*		*Corresponding position of geomagnetic poles*
		Declination	*Inclination*	
PRE-CAMBRIAN	600	N 56° W	− 38	N: Mid-Pacific 130° W, 0° N
(Torridonian)	—	S 69° E	+ 19	S: Near Ethiopia 50° E, 0° N
PALAEOZOIC				
Cambrian ..	450	S 11° W	+ 41	N: Marshall Islands 170° E, 15° N S: South Atlantic 10° W, 15° S
Silurian ...	350	S 25° W	− 16	N: North Honshu, Japan 140° E, 40° N S: South Atlantic 40° W, 40° S
Devonian ..	300	N 34° E	+ 2	N: Near Kamchatka 155° E, 45° N
		S 18° W	− 2	S: South Atlantic 25° W, 45° S
Permian ..	200	S 19° W	− 9	N: Near Kamchatka 155° E, 45° N S: South Atlantic 25° W, 45° S
MESOZOIC				
Triassic ..	180	N 26° E	+ 28	N: Near Kamchatka 155° E, 48° N
		S 34° W	− 28	S: South Atlantic 25° W, 48° S
TERTIARY				
Eocene ..	60	S 14° W	− 60	N: New Siberian Islands 133° E, 76° N S: Antarctica 47° W, 76° S

polar wandering assumes no relative motion of the land masses, but only a shift of the Earth relative to its axis of rotation in space. The modern theory of continental drift originated with Wegener, who supposed that in late Mesozoic times the continents drifted apart, having previously been grouped together in two masses called Laurasia and Gondwanaland. The theory is supposed to explain the following curious facts:

1. The similarity of the west coast of Africa and the east coast of South America.

2. The supposed similarities of fauna and flora in those parts of (*a*) North America and Europe and (*b*) South America, Africa, India, and Australia, which would have been joined together as the original two continental masses of Laurasia and Gondwanaland.

3. The glaciations of Permian and Carboniferous times, which were widespread throughout Gondwanaland.

The theory of continental drift has had a great fascination for certain scientists: no symposium on the subject fails to attract great attention. It is, however, based on no unambiguous geological evidence, and the geophysical case against it (H. Jeffreys [15]) has never been answered. Its plausibility arises principally from the attractive conception of the continents as rafts floating on a basaltic substratum, like icebergs floating in the sea. This analogy has, however, one fatal flaw: the strength of the continental rafts is less than that of the basaltic substratum under the oceans. The wide popularity of the theory of continental drift seems to be due to a feeling that it must somehow be simpler to move the continents than to move the Earth relative to its axis of rotation in space.

Perhaps the most unambiguous geological evidence relating to earlier positions of the land masses is that deduced from former climates. Yet in discussing this with reference to the above theories, geologists time and time again refuse to admit the possibility of polar wandering. The best-known climatic zones are those of the Carboniferous. In the latter half of last century beds of tillite—the boulder clay characteristic of glaciations—were found in South Australia, India, South Africa, and South America. That of Australia is of Permian age, those in India and South Africa are Permo-Carboniferous, and the South American glaciation is early Carboniferous. They are not contemporaneous glaciations—perhaps many tens of millions of years separate them. Yet it is often argued that polar wandering cannot explain this widespread glaciation and that we must invoke the idea that these four continents were at that time close together. In the same geological epoch the great coal forests were flourishing in North America, Europe, Russia, and China. Beds of laterite and bauxite, which now form only in the humid tropics, are also found in the Upper Carboniferous in these places. It is, of course, possible that these changes of climate may

have affected the Earth as a whole and may have resulted from fluctuations of solar radiation. The establishment of contemporaneity of geological age is perhaps not good enough absolutely to exclude this possibility.

It is of interest to examine the results of measurements of the magnetization of Carboniferous rocks. J. C. Belshé, of the Department of Geodesy and Geophysics, Cambridge University, has recently shown that the Carboniferous strata of Derbyshire and Yorkshire are magnetized with a low angle of dip, as if they had been magnetized in a field appropriate to the equatorial belt. Similarly D. I. Gough [16], of the Bernard Price Institute for Geophysical Research, University of the Witwatersrand, has recently shown that the Pilansberg dykes near Johannesburg are magnetized nearly vertically, as if the north magnetic pole had been in Abyssinia when they were formed. Though the age of these dykes is rather doubtful, they are earlier than late Carboniferous but probably later then Ordovician.

It will be possible in the next few years to see whether the hypothesis of polar wandering provides a satisfactory general explanation of the pre-Tertiary palaeomagnetic directions, by thoroughly comparing rocks from different continents. The present writer believes that this, and not continental drift, as Blackett [7] seems to suggest, will prove to be the key to the magnetism of rocks. We may not have long to wait before the matter is settled, but in any event a new and exciting chapter in the Earth's history seems to be on the verge of disclosure.

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Book reviews

HISTORICAL SCIENTIFIC BOOKS

Scientific Books, Libraries and Collectors, *by John L. Thornton and R. I. J. Tully. Pp. X + 288. The Library Association, London.* 1954. *24s. net.*

This work is a companion volume to 'Medical books, libraries and collectors,' published in 1949. The principal part of the work consists of an account of the more important scientific books up to and including those of the nineteenth centuries. These are classified first by date, then by subject and author. The book is not a formal bibliography, but gives a readable account of the authors and their books. It is obviously very far from exhaustive, and every historian of science will no doubt think that his own subject has been inadequately treated: the reviewer, for example, finds the account of alchemy very slight; it is surprising, too, to find that the magnificent 'Pirotechnia' of Biringuccio and Lazarus Ercker's work on assaying are missing.

The authors are, however, too modest in stating that the professional historian of science will find in their pages little that is new: such a historian, outside his special subject, will find the book of great use, and it will receive a permanent place on the reviewer's table. In addition to the general survey of scientific books there is a good account of scientific bibliographies, and also chapters upon scientific societies and libraries, all valuable. There is some inconsistency in the transcription of names, which are often half Latin and half Greek. Thus we have both Timaeus and Timaeos but not Timaios; Albertus Magnus sometimes appears, rather surprisingly in an English book, as Albert der Grosse. There are also a good many misprints, which is unfortunate in a book from which much will be copied. F. SHERWOOD TAYLOR

CHAUCERIAN ASTRONOMY

The Equatorie of the Planetis, a manuscript treatise ascribed to Chaucer; *edited by D. J. Price, with a linguistic analysis by R. M. Wilson. Pp. xvi + 214. Cambridge University Press, London.* 1955. *52s. 6d. net.*

Though the history of science is still far too neglected a subject, the historian can have not more than a faint hope of

unearthing an overlooked scientific treatise of major importance. This good fortune has, however, fallen to the lot of Dr Price—and deservedly so, for his careful scholarship had already won recognition. When examining manuscripts in the library of Peterhouse, Cambridge, he observed that in an English text on an astronomical subject the date 1392 frequently occurred, and at first surmised that it might be a missing part of Chaucer's 'Treatise on the Astrolabe,' written in 1391. Closer investigation proved that it was in fact a description of an equatorium or planetary calculator, of which only one medieval example appears to have survived, namely that at Merton College, Oxford.

This discovery was exciting enough in itself, but the excitement grew when Dr Price found the signature 'Chaucer' in a marginal note on one of the folios of the manuscript. There was at least the possibility that the treatise was not only by Chaucer but in his holograph, and this possibility is discussed at length in one chapter of the book. While not claiming to have definitely proved the point, Dr Price adduces sufficient evidence to make it highly probable that in 'The Equatorie of the Planetis' we have an original work by Chaucer written by his own hand: the find is therefore not only of scientific interest but of very considerable literary value.

In this edition the text is reproduced in facsimile and is accompanied by a transliteration and a translation into modern English. The critical apparatus includes a history of the equatorium and an account of medieval astronomical theories. There is a glossary of unfamiliar terms, and Mr R. M. Wilson provides a useful linguistic analysis. As usual, the Cambridge University Press has done the work of production extremely well, in a style befitting the exceptionally interesting subject. To use a phrase now outmoded, this is a book 'for every gentleman's library.'

E. J. HOLMYARD

PLANETARY ASTRONOMY

Guide to the Planets, *by Patrick Moore. Pp.* 222. *Eyre and Spottiswoode (Publishers) Limited, London.* 1955. 21*s. net.*

The present state of knowledge about the planets of the solar system is adequately described in this volume. This knowledge is based upon visual, photographic, and spectroscopic observations, combined with theoretical considerations. The subjects of space travel, voyages to the planets, and the establishment of a Martian base are inevitably brought in. The author is a well-known amateur observer, and some of the views put forward are his own personal opinions and not universally accepted. He states, for instance, that the theory that the lunar craters were made by meteors is 'quite untenable,' and that no practical lunar observers have any use for it now. That is certainly not so. The importance of some of the observations of lunar features made by amateur observers is somewhat shaken by the very considerable divergences between drawings of the same features—made usually from observations with telescopes of very moderate resolution—and the interpretation of these observations. Some of the lunar observers appear, moreover, to be under the impression that one theory and one theory only should account for every type of lunar formation.

The volume will form a useful guide for the planetary observer, who should use his own judgment, however, in weighing the evidence for different points of view. Most of the recent work has been well summarized. The book is illustrated with some good photographs and with some excellent coloured drawings by L. F. Ball.

H. SPENCER JONES

METEOR ASTRONOMY

Meteor Astronomy, *by A. C. B. Lovell. Pp. xiv* + 463. *Oxford University Press, London.* 1954. 60*s. net.*

Since the 1930's our knowledge of meteors has been revolutionized by modern photographic methods and the spectacular radio-echo techniques. In this comprehensive survey the author, himself a leader in the field of radio-astronomy, places the remarkable period of transition and development in its true perspective in relation to the long patient work of the past. In this way Professor Lovell has forged a vital link in the technical literature of meteors. Whether one is seeking information about technique, a critical and lucid exposition of the work of individual observers, or an objective account of the data of observation and the present state of meteor theory, this will be the book to turn to now and for many years to come.

The thoroughness and detail of the treatment have necessitated the complete exclusion of any discussion of meteorites or the physics of meteors; but we look forward to the adumbrated companion volume on these topics which, while not strictly part of meteor astronomy, are part of that 'question mark' in the human mind that first prompted the scientific observation of 'shooting stars.'

P. J. TREANOR

PHYSICAL CALCULATIONS

Advanced Level Examples in Physics, *by R. V. Johns and W. F. Ware. Pp.* 346. *Macmillan and Company Limited, London.* 1954. 16*s. net.*

This collection of some thousand examples, covering the usual courses in physics for the advanced level of the General Certificate of Education, differs from most other similar books in that the subject is divided into chapters, and that in each chapter the examples for practice are preceded by a brief theoretical introduction and some worked examples. Even though these introductions are often so brief as to degenerate into collections of formulae, these special features nevertheless add to the usefulness of the book.

Unfortunately the book is not as good as it might be. In general it barely reaches advanced level standard, and at some points it falls short of it. Subjects omitted completely include the method of dimensions, polarization, adiabatic expansions, self- and mutual inductance, and lines and tubes of force; there is no theory or worked examples on the wave theory of light nor on dielectrics. This list is by no means exhaustive.

But perhaps the most serious fault of the book is that there is no reference whatsoever to modern developments, in either the theoretical introductions or the worked examples. To give but one example—electrons are not mentioned at all. This is not the way to encourage the inquiring spirit in young physicists.

L. R. B. ELTON

STRESS AND STRAIN

Analysis of Deformation, *Volume* 2, *by K. Swainger. Pp. xxxvi* + 365. *Chapman and Hall Limited, London.* 1954. 70*s. net.*

The first nine chapters of this second volume are concerned with theoretical and some experimental aspects of stress and strain. Chapter X (the best) treats the deformation of rubber, chapter XI the application of couples, and chapter XII the shearing of thin plane laminae. The author states that the book is concerned primarily 'with practical aspects . . . without unduly emphasizing the mathematics.' The reviewer finds that

this is not so. For example, too much space is occupied with an explicit statement of the components of stresses in various co-ordinate systems, and general formulae abound on most pages. Some particular problems are worked out in detail, as in chapter XI; it is a pity that there is not more of this and less of generalities.

The book has been written because 'it proved impossible to have published a full account in the scientific journals.' This is, in the opinion of the reviewer, insufficient justification. It is noted that the book deals largely with matters which are controversial rather than with material which is widely accepted, and in many places the reader is left to form his own opinion. This situation is unusual in books on physics and engineering, and it is felt that such books should be reserved only for the publication of material which is reasonably firmly established. The book has what might be called a 'microstructure,' which is unnecessary. For about 350 pages of text there is a contents list occupying no less than sixteen pages, and the index runs to eleven pages. The explanation is the abundance of sub-headings, there being, for example, no fewer than seven of these on page 141. Six pages are used to state the principal notation, and the preface occupies eight pages. All this, together with the almost unnecessary mathematical appendix, goes to make what is an expensive volume.

L. S. GODDARD

CRYSTAL LATTICE THEORY

Dynamical Theory of Crystal Lattices, *by Max Born and Kun Huang. Pp. viii + 420. Oxford University Press, London.* 1954. 50*s. net.*

Almost forty years ago, Professor Born published the first detailed account of this subject. In the intervening years he has been largely responsible for several reports making the theory up to date, but the latest of these is now twenty years old. In collaboration with Professor Huang he has now prepared the present volume, which is a new and comprehensive treatment and will undoubtedly remain authoritative for many years to come.

The subject of lattice dynamics suffers from the difficulty of sheer mathematical bulkiness. Most of the calculations are simple enough in principle, and may readily be written down for the case of a monatomic Bravais lattice, but the general formulae necessary to describe real physical systems are usually very unwieldy, involving summations over numerous indices and many different terms. The authors have made a considerable effort to achieve conciseness of notation; even so, expressions involving six or seven lines of type are not uncommon, and there are moments when one feels that one would like to be able to see more easily through the formalism to the basic algebra and the underlying physical ideas.

For this reason it is perhaps fortunate that we have here, in effect, two books under one cover. The first three chapters, written mainly by Professor Huang, deal in an elementary way with atomic forces, vibrations, and the elasticity and stability of lattices. The physical aspects are emphasized, and relevant experimental results are discussed. Many topics are reviewed, yet the argument has a satisfying unity which makes it easy to read, even for the nonexpert.

The second part (which represents the original nucleus of the book, as prepared by Professor Born) invokes full mathematical rigour; we are led from a quantum-mechanical assembly of nuclei and electrons, through the complexity of the method of long waves, to general formulae for the piezo-electric, pyro-electric, and optical phenomena. The detailed derivation of these formulae (many published here for the first time) will be of the greatest value to the specialist, and the ten appendices contain some elegant notes on the mathematical techniques used in the theory.

It may well be that the first part will be more widely read today; the rigorous treatment may prove the more permanent contribution. J. M. ZIMAN

ORGANIC SYNTHESIS

Organic Syntheses, *Volume 34, edited by W. S. Johnson. Pp. vi + 121. John Wiley and Sons Inc., New York; Chapman and Hall Limited, London.* 1954. 28*s. net.*

Most organic chemists are already familiar with this series and will welcome this latest member. The present volume lists the preparations of thirty-six compounds, of which several are prepared in two or three stages, so that the actual number of preparations described is nearer fifty. As in the previous volumes, complete details are given of manipulation, techniques, sources of reagents, and any necessary precautions, so that no unforeseen difficulty should arise in repeating the experiments. In addition, notes on the preparation of the α:ω-dinitro derivatives of propane, pentane, and hexane are appended to that for 1:4-dinitrobutane.

Some of the compounds here described, such as *cyclo*heptanone and di-*tert*-butyl malonate, are clearly of importance as intermediates; others, such as 3-methylthiophen and azelanitrile, have been included mainly because they illustrate general reactions and procedures. The index is cumulative and covers volumes 30–34.

The high standard of previous volumes is maintained, and the present volume is a worthwhile addition to the series. J. F. W. McOMIE

ORGANIC CHEMISTRY

Outlines of Organic Chemistry (*third edition*), *by E. J. Holmyard. Pp.* 492. *Edward Arnold (Publishers) Limited, London.* 1954. 16*s. net.*

Dr Holmyard is well known for his skill in using his wide knowledge of the history of chemistry for both enlivening his textbooks and making them more easily understood and memorized. In this book the author continually stresses the contributions made by individuals to the development of chemistry. A study of the impact of the personality of an investigator on his work is an inspiring theme and one of the best ways of introducing a student to the history of any science and of arousing his enthusiasm.

The question of how much 'special' work should be included in a book of this kind has, on the whole, been solved very satisfactorily, as indicated by the treatment of physical properties, stereoisomerism, tautomerism, valency, free radicals, and catalysis. By omitting the quite unnecessary questions after each chapter useful space could have been saved and utilized to provide a more extensive treatment of aromatic substitution. Moreover, work in this field has now gone too far to be dismissed as 'attempts to explain the direction of substitution.'

The reference to nitronium and nitracidium ions is, however, welcome and the emphasis laid on the natural occurrence of simple organic compounds will be most stimulating to all who are attracted by the impingement of chemistry upon other sciences.

F. CHALLENGER

PORPHYRINS

Porphyrins, their Biological and Chemical Importance, *by A. Vannotti (translated by C. Rimington). Pp. xv* + 258. *Hilger and Watts Limited, London.* 1954. 50*s. net.*

Professor Vannotti explains in his foreword that the book is not 'a mere revision' of his earlier monograph (*Porphyrine und Porphyrinkrankheiten*, 1937). While the present book contains considerable extra material, which enhances its value over that of the 1937 monograph, too many of the arguments presented seem to take little or no cognizance of important work done since 1945 on porphyrin metabolism, largely with the aid of isotopic tracers. References to such work are made, but they are not adequately woven into the material presented. This has inevitably led to contradictions. Professor Vannotti appears, in fact, reluctant to part with his earlier views. For example, he states (p. 88) 'the conclusion seems warranted that the liver regularly transforms protoporphyrin into coproporphyrin . . .'; yet, in a brief summary (p. 31) of the studies by American and British workers using isotopes, is set out the opposite, and now generally held, view.

Clinical aspects of the various porphyrias are discussed fully and well; there are excellent descriptions of the symptoms, supported by case histories. The author's preoccupation with lactoflavin therapy is somewhat misleading, since its efficacy is by no means assured.

The book is well translated; it is, however, unfortunate that publication has been so long delayed. J. J. SCOTT

PREPARATIVE ORGANIC CHEMISTRY

Methoden der Organischen Chemie (Houben-Weyl), *4th revised edition, edited by Eugen Müller. Georg Thieme Verlag, Stuttgart. Volume II:* Analytische Methoden. *Pp. xxiv* + 1070. 1953. *DM* 139 *net. Volume VII, part* 1*:* Sauerstoffverbindungen II, Aldehyde. *Pp. xxiii* + 556. 1954. *DM* 82 *net. Volume VIII:* Sauerstoffverbindungen III. *Pp. xviii* + 776. 1952. *DM* 98 *net.*

The third edition of this compendium was esteemed second only to Beilstein in the library of the bench-working organic chemist, and it is safe to say that the new fourth edition, completely revised by an authoritative group of scholars under the leadership of E. Müller and supported by O. Bayer, H. Meerwein, and K. Ziegler, will take an even more definite place as a real essential for the investigator.

The main part of the work is to be devoted to methods of preparation which will be classified as general methods ranging over several groups, and also as the methods applicable to selected groups of organic compounds. Two volumes exemplifying the latter method of approach have now appeared, namely Volume VII, part I, oxygen compounds II (1954), and Volume VIII, oxygen compounds III (1952). The author of the former is Otto Bayer, who deals with the aldehydes in an exhaustive manner which is altogether praiseworthy. The special feature which marks the work is the profusion of examples of preparations and also the very full reference to the original literature, including that relating to patents. A greater stimulus to research than this compilation could hardly be imagined. Every organic chemist has been, is now, or will be, interested in aldehydes, and Dr Bayer tells us exactly what we want to know about them. It is one of the most intriguing monographs which the writer has been privileged to read.

The reviewer was specially interested in R. Criegee's article on the organic peroxides in the second of these two volumes. Though this, and probably all other sections, could not be regarded as exhaustively treated, there is very much that is both unfamiliar and suggestive of new ideas in this description of a field of growing industrial importance. The new Houben-Weyl makes its own characteristic contribution to our resources and does not clash with those other most valuable works of reference, 'Organic Syntheses,' 'Inorganic Syntheses,' and 'Organic Reactions.' Indeed, one is glad to note a considerable number of references to these series.

Volume II (1953) deals with analytical methods, both qualitative and quantitative. This volume of over a thousand pages has been compiled by some twenty-five contributors, and it appears to be a moderately complete account of the parts of the subject treated. It must be regarded as a work of reference; it is obviously a mine of information on available techniques. The ordinary organic chemist will browse most happily in the second section of part I, which deals with the determination of the most common functional groups.

This enterprise deserves the warmest encouragement, and the purchase of the parts as they appear is obligatory for scientific libraries serving schools of research, whether they be in universities or in industrial or government organizations. ROBERT ROBINSON

INORGANIC CHEMISTRY

Inorganic Chemistry (*sixth edition*), *by Fritz Ephraim. Edited by P. C. L. Thorne and E. R. Roberts. Pp. xii* + 956. *Oliver and Boyd Limited, Edinburgh.* 1954. 35*s. net.*

Unlike many textbooks, Ephraim improves with age. There is, as a matter of fact, not a great deal of difference between this edition and its predecessor of 1948. The general character of the book remains unaltered, and the prefaces to the two editions are almost identical. A number of new sections, important in their bearing on chemistry but concisely inserted into the old framework, succeed in bringing the text into line with modern developments without burying established principles of the subject under a mass of detail put in merely for the sake of its newness. Recent discoveries in the transuranic series of elements, a brief account of the clathrate compounds, and new work in the field of metal hydrides are naturally among the items so introduced. But the fact that the bulk of the text stands without the need of more than minor modification, in spite of the relatively enormous output of new factual material in the inorganic field, and also that the new items can be fitted in so conveniently without disturbing the general run of the text, shows that the original selection of basic material in the earlier editions was well made and that the arrangement lends itself to the process of making up to date.

The main revisions have been made in the fields of radioactivity and valency theory; in the latter the introduction of three pages on 'quanticule theory' is perhaps the most original feature of the edition. In the earliest editions it was not always easy to find what one wanted, but the pruning and revision necessary for the production of subsequent editions of not greatly increased size have resulted in a greater clarity, which is maintained in this edition also. Although they were good in the fifth edition, the actual printing and setting out of the text have been further improved; this makes the book pleasant to handle and read, while the price, although slightly increased, remains very moderate indeed. F. M. BREWER

FLUORESCENCE

Fluorescence Analysis in Ultra-violet Light (*fourth edition*), *by J. A. Radley and J. Grant. Pp.* 560. *Chapman and Hall Limited, London.* 1954. 52*s.* 6*d.*

Fluorescence has something of magic in its appearance, eludes easy estimation, and has a very large measure of empiricism in its use. The subject has two sides, that of the fluorescence of highly purified chemical substances, from which alone a connection can be made with physical and chemical theory, and that of materials as they are usually found. Among pure chemical substances fluorescence is uncommon; in organic chemistry it is chiefly confined to molecules with certain types of ring structure. Materials such as paper, oils, fats, minerals, drugs, skin, and so on frequently show fluorescence to some degree, almost always due to minute quantities of fluorescent substances present with the main chemical components.

The fourth edition of this book, revised, partly rewritten, and with 'a good deal of dead wood cut out,' deals with the second aspect of the subject, and provides a wealth of details and references on the use of fluorescence observations over a wide range of applied chemistry. With the growth of the subject that wealth has become embarrassing, and the impression received by a newcomer may prove bewildering. The subject still remains one where individual experience coupled with the closest scientific scrutiny is the only way to avoid gross errors. E. J. BOWEN

MOLECULAR THEORY

Molecular Theory of Gases and Liquids, *by J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird. Pp. xxvi* + 1219. *John Wiley and Sons Inc., New York; Chapman and Hall Limited, London.* 1954. 160*s. net.*

This is the most comprehensive account yet published of the molecular interpretation of the main physical properties of gases and liquids. The authors' own experience at the University of Wisconsin has been supplemented by the collaboration, on specific topics, of other experts, including J. de Boer of Amsterdam (quantum effects) and J. S. Rowlinson of Manchester (polar gases). The book ranges from fundamental theory and its detailed comparison with experiment, to illustrative examples and numerical problems, in a manner which should ensure its becoming a standard work of reference.

Apart from some preliminaries on classical, quantum, and statistical mechanics, two main sections deal with the equilibrium properties exemplified by the equation of state and with the transport phenomena of viscosity, diffusion, and thermal conductivity. Consistent use is made of the generalized principle of corresponding states in summarizing experimental data, which are then interpreted in terms of the various empirical functions for the potential energy of a pair of interacting molecules. A concluding section reviews our much less complete understanding of the theory of intermolecular forces. Chemical reactions are not specifically discussed, though a chapter on hydrodynamics includes flames, shock waves, detonations, and rocket propellents.

A valuable appendix provides tables by means of which the experimentalist or engineer can quickly estimate from fundamental constants many unknown quantities of practical importance.

K. W. SYKES

ORGANIC REACTIONS

Organic Reactions, *Volume* 8, *edited by Roger Adams. Pp. v* + 437. *John Wiley and Sons Inc., New York; Chapman and Hall Limited, London.* 1954. 96*s. net.*

The appearance of a new volume of 'Organic Reactions' is always an important event for organic chemists. The project, due to Professor Roger Adams, of compiling critical discussions by recognized authorities of the more important organic reactions first bore fruit in 1942.

The value of these books to the chemist is threefold. In the first place, he is sure of a comprehensive and critical account of the scope and modus operandi of the reactions described. Secondly, he will find a comprehensive list of compounds prepared by the reactions described, together with references. Finally, particularly in the more recent volumes, there are stimulating discussions of reaction mechanisms. A readable, interesting style is preserved throughout.

The present volume, the eighth in the series, preserves the tradition of the earlier ones and will acquire the same esteem. It contains sections on catalytic hydrogenation of esters to alcohols; the synthesis of ketones from acid halides and organo-metallic compounds of magnesium, zinc, and cadmium; the acylation of ketones to form β-diketones or β-keto aldehydes; the Sommelet reaction; the synthesis of aldehydes from carboxylic acids; the metalation reaction with organolithium compounds; β-lactones; and the reactions of diazomethane and its derivatives with aldehydes and ketones. The admirable article on the acylation of ketones deals with reactions and compounds of great current importance and will be widely appreciated. The critical discussions on the mechanisms, particularly of the acid-catalysed cleavage of the Reissert compounds and of the Sommelet reaction, will also be welcomed. D. LL. HAMMICK

PHYSIOLOGICAL CHEMISTRY

Practical Physiological Chemistry (*thirteenth edition*), *by Philip B. Hawk, Bernard L. Oser, and William H. Summerson. Pp. xvi* + 1439. *J. and A. Churchill Ltd., London.* 1954. 85*s. net.*

The present edition of 'Practical Physiological Chemistry' justifies in every way Dr Hawk's pride in his achievement in producing thirteen editions of the book in the course of the last fifty years. The esteem which the book has gained lies not only in the range and detail of its treatment of biochemical methods but in the freshness which revision has infused into each edition by the inclusion of the newest developments in biochemistry. Taken together, the several editions would provide interesting material for a study of the growth of biochemistry over the last half-century.

In its essential plan the present edition conforms to that of the more recent ones, except that theoretical aspects have gained greater recognition. In addition to descriptions of the conventional and established methods for the study of body tissues, fluids, and secretions which have made it an invaluable aid to teachers and clinical biochemists, the book includes description of the new techniques of biochemistry—ion exchange, counter-current distribution, paper chromatography, liver function tests, and so on. The vigour of the revision for this edition is well illustrated by reference to such matters as the helical structure of proteins, cobalamins, and thioctic acid, the succinct accounts of which many of the less academic biochemists will find helpful. Equally valuable are the numerous sections that deal with matters of clinical importance. Dr Hawk and his co-editors and collaborators can be warmly congratulated for this impressive new edition.

J. R. P. O'BRIEN

MINERALOGY

Rocks and Mineral Deposits, *by Paul Niggli. (English translation by R. L. Parker.) Pp. xiii + 559. W. H. Freeman and Company, San Francisco; Bailey Bros. and Swinfen Limited, London.* 1954. 102*s. net.*

This book is for the student or research worker concerned with rocks and minerals; it would be of only limited value to other scientists wishing to learn something of these things. It follows an unconventional plan, but since the author was one of the great petrologists of this century it is of value to see the data presented in this way. The German edition was published in 1948, and this able translation by Professor R. L. Parker was done under the direct supervision of Professor Niggli, who was able to write a preface but died before the translation appeared.

An early section, and later a whole chapter, is devoted to the author's method of calculation of chemical analyses of rocks into standard minerals, and their graphical presentation. The book provides the most readily accessible explanation in English of this useful method. Whenever possible the author makes a quantitative approach to the subject with which he is dealing. He also makes a brave attempt to give precision to the description of the fabric of rocks. Only limited parts of his methods and ideas are likely to be generally assimilated into petrology, and in fact the author himself makes little use of these in his final chapters on a classification of the rocks.

L. R. WAGER

TRACE ELEMENTS IN SOILS

Geochemie seltener und nur in Spuren vorhandener chemischer Elemente im Boden, *by A. P. Winogradow. Pp. x + 250. Akademie-Verlag, Berlin.* 1954. *DM 23 net.*

Winogradow's book, published in Moscow in 1950, was the first comprehensive discussion of the distribution of trace elements in soils, and this German translation by Nikolai Ziabkin is welcome, as it makes the information much more readily accessible. The proportions of over thirty elements in some twenty Russian soil profiles, covering the major world soil groups, are discussed, and compared with values reported in the literature. In the main, only total contents are considered, and there seems to be a tendency to generalize on rather insufficient data. Following a detailed consideration of individual elements, the general principles applicable to different soil types are summarized in the final chapter. A few of Winogradow's ideas on the normal distribution of trace elements may give rise to some discussion, particularly among workers investigating soils which are somewhat younger and less fully developed than those of the Russian plains.

The translation has been rather uncritical, as many obvious numerical errors in the original are repeated without correction. Most of these are due to the unfortunate use of percentages rather than parts per million to indicate soil contents; 15 p.p.m., for instance, is expressed as $1{,}5.10^{-3}$ per cent, and mistakes often occur in the exponent. The reviewer has noted numerous such slips, and consequently doubt arises over the authenticity of other values which are not so readily checked. Misspellings of authors' names and misquotations of references are also rather abundant; nevertheless the book is a valuable addition to soil literature.

R. L. MITCHELL

GEOLOGY OF PETROLEUM

Geology of Petroleum, *by A. I. Levorsen. Pp. x + 703. W. H. Freeman and Company, San Francisco; Bailey Bros. and Swinfen Limited, London.* 1954. 68*s. net.*

A book on the geology of petroleum by A. I. Levorsen is an event, and those of us who were aware of its imminence looked forward to a treatise both comprehensive and authoritative. In this we have not been disappointed, for this volume covers every aspect of petroleum geology. Within its pages there is a vast array of factual information, brought together so admirably that it sheds light on every conceivable aspect of what has now grown to be the widest field of applied geology. Each chapter, moreover, includes a well-stocked bibliography to guide the student to a wider range of reading.

The author regards his subject primarily from the practical angle. To him, the essence of the duty of a petroleum geologist lies in the quest for more oil, and therefore he begins from the very outset to describe what are the essentials of an oil pool, and how its external and internal structure vary according to the natural forces which brought it into being. His sections dealing with the structural accumulation of oil and gas, and notably his chapter on the stratigraphic factors governing the accumulation of petroleum, are the highlight of the volume.

Later sections consider the relationship of water, oil, and gas, within the reservoir rock, followed by chapters devoted to sub-surface geology, petroleum provinces, and petroleum prospects.

The book will be welcomed by all students of the subject as an outstanding work.

V. C. ILLING

HISTOLOGY

Trattato di Istologia (*fourth edition*), *Volumes 1 and 2, by Giuseppe Levi. Pp. xvi + 1171. Unione Tipografico-Editrice Torinese, Turin.* 1954. 12 000 *lire net.*

The fourth edition of this textbook by the senior Italian histologist appears eight years after the third edition (1946). In line with the tradition of the German and Italian schools of medicine, it deals exclusively with the structural characteristics and biological properties of cells and tissues and not with the structure of the various organs; the only relevant exception is the structure of blood vessels, discussed in chapter 31.

The general pattern and guiding principles are basically the same as in previous editions. However, many important changes have been introduced and a good number of illustrations substituted or added. A concise review of the histochemical and histophysical techniques and of the tools for the investigation of microscopical and ultramicroscopical structures has been included. Mitosis, meiosis, the chromosomes, and the ultrastructure of the cytoplasm in general and of the various kinds of cells, have been dealt with more extensively than in the third edition.

Though the author gives his personal views on the various topics, facts, and opinions deriving from the contributions made by biologists of other countries are reported and objectively discussed throughout. No dogmatic statements, which often may seem inevitable in treatises written chiefly to meet the needs of students, are to be found. Professor Levi's book represents an invaluable guide also for the specialist histologist.

The list of references, set under thirty separate headings at the end of the second volume, will not only serve as an introduction to a more detailed bibliography but is in itself a rich source of information. A detailed subject index and an author index are also included. This very original book once again teaches the morphologist that the old

and seemingly impenetrable barriers between neighbouring fields of biology must be broken down in order to get a deeper knowledge of living matter.

R. AMPRINO

BIOLOGICAL KINETICS

The Kinetic Basis of Molecular Biology, *by F. H. Johnson, H. Eyring, and M. J. Polissar. Pp. vii + 874. John Wiley and Sons Inc., New York; Chapman and Hall Limited, London.* 1954. 120*s. net.*

There can be no doubt today about the importance of the borderland between chemistry, physics, and biology, and this book is an important contribution to its study. The unifying theme is the application to a series of biological phenomena of principles and ideas derived from the theory of chemical reaction rates.

The book contains an immense amount of interesting and valuable material, and will be of interest and assistance to those who begin with some knowledge of the various specialized topics with which it deals, for example luminescence, membrane diffusion, muscle contraction, the nerve impulse, and drug action. In fact, for certain topics it constitutes something approaching a small reference work.

If it is true that a good many biologists chose their subject because they disliked mathematics they will find the first six chapters pretty tough going. These constitute in fact an advanced textbook of thermodynamics, and discuss quantum and statistical mechanics which most physical chemists will find demands their full attention. Then, on page 123, the book embarks upon a fairly elaborate review of the subject of luminescence, and follows with reviews of other subjects, in no very obvious order.

If a general criticism is to be made, it is perhaps that the liaison between the physico-chemical and the biological sides is not after all really very clearly brought out. It is perhaps most successfully achieved in chapters such as that on the effects of temperature, where numerous biological effects are treated under one physico-chemical heading.

For some interested in the very general kind of question discussed for example in Schrödinger's 'What is Life?', there may be a certain disappointment, especially if the name of at least one of the authors had led them to expect some bold and uninhibited reflections on these fundamental matters. But there is no reason for real complaint that the authors have chosen to plan their book as they have, and there is no doubt of its value.

C. N. HINSHELWOOD

MARINE BIOLOGY

Aspects of Deep Sea Biology, *by N. B. Marshall. Pp.* 380. *Hutchinson's Scientific and Technical Publications, London.* 1954. 35*s. net.*

'. . . contemporary science can be extremely beautiful though often very exasperating, and at times tremendous fun.' This quotation, cited by the author in his preface, is not only substantiated by the contents of the book itself, but may also account for the patently sincere and engaging style in which it is written. The work takes the form of a digest—a compendium of short and more or less self-contained articles, each dealing with a particular aspect of deep-sea biology, but skilfully arranged in sequence to constitute an orderly, well balanced, and comprehensive review of the subject as a whole. In keeping with this method of presentation, the many beautiful illustrations by the author's wife are identified by the chapter number of the text to which they relate; at the end of each chapter references are given to the authorities quoted. This book is specially worthy of commendation to the biological student as a sound and stimulating introduction to our knowledge and concepts of life in the ocean deeps, as revealed by scientific investigation since the world voyage of H.M.S. *Challenger* from 1872 to 1876. It can be equally commended to the intelligent lay reader for its informative and compelling reading.

E. FORD

INSECT METAMORPHOSIS

The Physiology of Insect Metamorphosis, *by V. B. Wigglesworth. Pp.* 152. *Cambridge University Press, London.* 1954. 12*s.* 6*d. net.*

Insect physiologists will be indebted to Professor Wigglesworth for this incisive survey of the progress that has been made in the study of insect metamorphosis. The rapid growth of interest in this field is well shown by the fact that of over four hundred publications reviewed almost one-quarter have appeared within the past five years. This monograph will be welcomed by both specialist workers and general readers as a timely statement of the present position.

In making his own well-known studies on the hormonal control of metamorphosis in *Rhodnius* its central theme Professor Wigglesworth has given his book authority and clarity. In regarding metamorphosis not as an extension of embryological development but as an expression of polymorphism he has given both renewed support to a theory due to Swammerdam and a stimulus to further inquiry.

In writing the first of a new series of Cambridge monographs in experimental biology the author has set a high standard which will cause further contributions to the series to be eagerly awaited. The monograph is well illustrated and its production is of the quality one has come to expect from the Cambridge University Press.

R. DENNELL

BOWER-BIRDS

Bower-Birds, *by A. J. Marshall. Pp.* 208. *Clarendon Press, Oxford.* 1954. 30*s. net.*

This book was much needed, for while bower-birds have often featured in discussions on evolution and behaviour, the facts and interpretations usually presented include many errors. After an introduction and two brief (but difficult) chapters on sexual physiology in birds, there follow eighteen chapters, each giving a straightforward factual account of one species of bower-bird, with special reference to its displays, bower, breeding, and development of sex organs. There is much first-hand observation and a good summary of the observations of others. A final chapter considers the evolution of bower-building.

This is a trustworthy account of what happens, and the book will therefore be of value to zoologists and also, one hopes, to amateur ornithologists, even though the author has unfortunately not attempted to simplify the language or presentation and introduces many technical physiological terms without definitions. But the tale that is told is astonishing. The bowers are of several patterns, which help in the classification of the species. The objects brought to them may be of restricted colours, which match colours on the female. The bowers may be stained—one of the few examples of the use of tools by birds. Some species are vocal mimics. The group offers a wonderful opportunity for future research.

D. LACK

ENGINEERING

Dizionario d'Ingegneria, *Volume* 4 MOT–RUL, *edited by E. Perucca. Pp. viii* + 1040. *Unione Tipografico-Editrice Torinese, Turin.* 1954. 12 000 *lire net.*

As this impressive undertaking approaches completion it becomes increasingly evident how much care and forethought have gone into its preparation and how thorough the editor has been in collecting all terms relating not only to engineering but to all cognate subjects.

The first three volumes have already been reviewed in ENDEAVOUR with abundant praise and only minor criticisms, which apply also to this fourth volume.

As the pattern unfolds, the system adopted for sub-headings and cross-references appears more and more a complete success: one is reminded of the innumerable connections and ramifications of the nerve cells in the brain which serve to transmit information rapidly and efficiently.

Certain entries are covered most thoroughly: for instance, the seventy-six pages given to the various types of motors could hardly be bettered. In other cases, however, one would like to learn more: for instance, there is little about modern developments in the plastics industry. But some unevenness in a work which is the combined effort of 125 experts is of course inevitable.

Some users, particularly outside Italy, may regret that the French, English, and German equivalents are given for only a few terms: a more generous allowance of these equivalents would in fact have made the 'Dizionario' also a technical dictionary in four languages.

This is a magnificent work of reference which will rarely fail to provide anybody consulting it with the required information. L. PINCHERLE

RARE METALS

Rare Metals Handbook, *edited by Clifford A. Hampel. Pp. xiii* + 657. *Reinhold Publishing Corporation, New York; Chapman and Hall Limited, London.* 1954. 96*s. net.*

This book is written by thirty-four contributors and contains chapters dealing with twenty-eight of the less common metals; the platinum metals and the rare earths are dealt with in single chapters. The book has many of the defects to be expected from such a wide joint authorship, and there is little or no general plan, except that the industrial methods of extraction and purification are described fully and a list of physical properties is given for each metal. Thus, the chapter on vanadium contains detailed tables of the properties of more than forty vanadium-bearing minerals, but for some metals the sources are only briefly summarized. The chapters are of very varying quality, but three general comments may be made. First, the main value of the book lies in the description of industrial methods for the extraction and purification of the metals, since this information is not easily accessible. Second, readers interested in the chemistry of the less common metals will be better advised to buy a standard textbook of inorganic chemistry, in which the facts will be seen as part of a general science. Third, the sections dealing with the present reviewer's interests—namely the properties and structures of metals and alloys—are sometimes inferior, unreliable, and out of date. Thus some lattice spacings are given in Ångström units when the values are really in kX units, while some values are given with no units indicated. Equilibrium diagrams of alloys are given for some metals only, and are often quite out of date. Any reader of the book will acquire much information, but it requires a thorough revision before it can be recommended as an authoritative source of numerical data.

W. HUME-ROTHERY

Some books received

(*Note.* Mention of a book on this page does not preclude subsequent review.)

ASTRONOMY

Astronautica Acta, Volume 1, *edited by F. Hecht. Pp.* 60. *Springer-Verlag, Vienna; I. R. Maxwell and Company Limited, London.* 1955. 62*s. net per annum.*

BIOLOGY

Actions of Radiations on Living Cells (*second edition*), *by the late D. E. Lea. Pp. xiv* + 416. *Cambridge University Press, London.* 1955. 30*s. net.*

Annual Report of the National Oceanographic Council 1953–54. *Pp.* 28. *Cambridge University Press, London.* 1955. 5*s. net.*

BOTANY

British Mosses and Liverworts, *by E. V. Watson. Pp. xvi* + 419. *Cambridge University Press, London.* 1955. 45*s. net.*

Embryogenesis in Plants, *by C. W. Wardlaw. Pp. ix* + 381. *John Wiley and Sons Inc., New York; Methuen and Company Limited, London.* 1955. 42*s. net.*

CHEMISTRY

Biochemistry of Nitrogen (in honour of Artturi I. Virtanen). *Pp.* 535. *Suomalainen Tiedeakatemia, Helsinki.* 1955. *Paper covers,* $10*; bound,* $12 *net.*

Catalysis, Volume 2: Fundamental Principles (Part 2), *edited by P. H. Emmett. Pp. vi* + 473. *Reinhold Publishing Corporation, New York; Chapman and Hall Limited, London.* 1955. 96*s. net.*

Chemical Process Principles: Part 1, Material and Energy Balances (*second edition*), *by O. A. Hougen, K. M. Watson, and R. A. Ragatz. Pp. xxxv* + 504. *John Wiley and Sons Inc., New York; Chapman and Hall Limited, London.* 1954. 68*s. net.*

The Chemistry of Micro-organisms, *by A. Bracken. Pp. vii* + 343. *Sir Isaac Pitman and Sons Limited, London.* 1955. 30*s. net.*

The Chemistry of Petroleum Hydrocarbons, Volume 1, *edited by B. T. Brooks, C. E. Boord, S. S. Kurtz, and L. Schmerling. Pp. viii* + 664. *Reinhold Publishing Corporation, New York; Chapman and Hall Limited, London.* 1954. 144*s. net.*

Les Gaz Inertes; l'Hydrogène — Les Halogènes, *by P. Laffitte and H. Brusset. Pp.* 396. *Masson et Cie, Paris.* 1955. *Paper covers, fcs* 3600*; bound, fcs* 4200.

An Introduction to the Electronic Theory of Valency (*third edition*), *by J. C. Speakman. Pp. vii* + 180. *Edward Arnold (Publishers) Limited, London.* 1955. 10*s.* 6*d. net.*

An Introduction to the Principles of Chemistry, *by L. H. Cragg and R. P. Graham. Pp. ix + 740. Clarke, Irwin and Company Limited, Toronto.* 1954. $7.50 *net.*

Journal of Inorganic and Nuclear Chemistry, Volume 1, Nos. 1–2, *edited by J. J. Katz, H. C. Longuet-Higgins, and H. A. C. McKay. Pp.* 164. *Pergamon Press Limited, London.* 1955. *Subscription A (normal)* 90*s. Subscription B (for subscriber's own use)* 70*s. per volume.*

A Manual of Paper Chromatography and Paper Electrophoresis, *by R. J. Block, E. L. Durrum, and G. Zweig. Pp.* 484. *Academic Press Inc., New York.* 1955. $8 *net.*

Monomeric Acrylic Esters, *by E. H. Riddle. Pp. vii* + 221. *Reinhold Publishing Corporation, New York; Chapman and Hall Limited, London.* 1954. 40*s. net.*

Photosynthesis, *by R. Hill and C. P. Whittingham. Pp. vii* + 165. *Methuen and Company Limited, London.* 1955. 8*s.* 6*d. net.*

Synthetic Rubber, *edited by G. S. Whitby. Pp. xii* + 1044. *John Wiley and Sons Inc., New York; Chapman and Hall Limited, London.* 1954. 144*s. net.*

GENERAL

Biology as a Career (*second edition*), *edited by G. E. Fogg. Pp.* 40. *The Institute of Biology, London.* 1955. 2*s.* 6*d. net.*

Composition of Scientific Words, *by R. W. Brown. Pp.* 882. *Published by the author at the U.S. National Museum, Washington.* 1954. $8 *net.*

Deterioration of Materials, *edited by G. A. Greathouse and C. J. Wessel. Pp. xvii* + 835. *Reinhold Publishing Corporation, New York; Chapman and Hall Limited, London.* 1954. 96*s. net.*

The Human Use of Human Beings (*revised edition*), *by Norbert Wiener. Pp.* 199. *Eyre and Spottiswoode (Publishers) Limited, London.* 1954. 18*s. net.*

The Scientific Journal of the Royal College of Science, Volume 24, *edited by B. W. Baker. Pp. vi* + 122. *Royal College of Science, London.* 1954. 10*s.* 6*d. net.*

Slovník Stavebné Mechaniky a Příbuzných Oborů v Pěti Řečích (Dictionary of Structural Mechanics, with keys in five languages), *by Z. Bažant. Pp.* 251. *Nakladatelství Československé Akademie Věd. Prague.* 1953. *Paper covers, Kčs* 15*; linen covers, Kčs* 20 *net.*

HISTORY OF SCIENCE

The Development of the Concept of Electric Charge: Electricity from the Greeks to Coulomb, *by Duane Roller and Duane H. D. Roller. Pp. iv* + 97. *Harvard University Press, Cambridge, Mass.* 1954. $1.60 *net.*

Oxford's Contribution to the Origins of Modern Science (a paper read to the British Association for the Advancement of Science at Oxford on 6th September 1954), *by A. C. Crombie. Pp.* 27. *Basil Blackwell, Oxford.* 1954. 2*s.* 6*d. net.*

Science and its Background (*second edition*), *by H. D. Anthony. Pp. ix* + 337. *Macmillan and Company Limited, London.* 1954. 20*s. net.*

MEDICINE

British Medical Bulletin, Volume 11, No. 1 (Blood Coagulation and Thrombosis), *edited by J. V. Dacie. Pp.* 82 + *xvi. The British Council, London.* 1955. 15*s. net.*

The Harvey Lectures (delivered under the auspices of the Harvey Society of New York 1953–1954). *Series XLIX. Pp. viii* + 299. *Academic Press Inc., New York.* 1955. $7.50 *net.*

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